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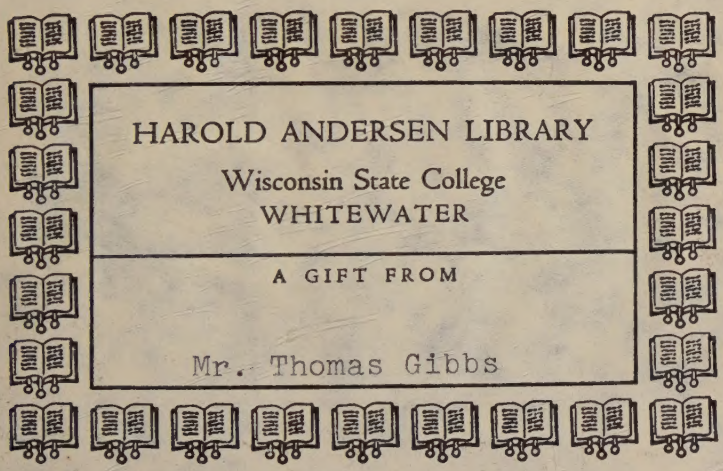
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THE PRODUCTION OF ALUMINUM AND ITS INDUSTRIAL USE.

P. S. TICE
DETROIT, MICH.

BY
ADOLPHE MINET,
Officer of Public Instruction and Editor of "L'Electrochimie."

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TRANSLATED, WITH ADDITIONS,

BY

LEONARD WALDO, S.D. (Ha[illegible]

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LEONARD WALDO.

AUTHOR'S PREFACE TO THE AMERICAN EDITION.

THE present work comprises a principal part, which is a literal translation of the German edition "Die Gewinnung des Aluminiums und dessen Bedeutung für Handel und Industrie," published in 1902, and an appendix including two wholly new chapters: the first, by the Author, is devoted to a supplementary consideration of those parts of the German edition which have been made the subject of criticism; in the second chapter, Dr. Leonard Waldo describes the developments in the Aluminum industry of recent years, more especially in the United States—a matter which the limited scope of the first book had compelled me, greatly to my regret, to overlook.

I am sure that Dr. Waldo's contribution will meet with a favorable reception on the part of the reader, and that his twofold collaboration (since he is also the translator) will contribute in large measure to ensure popular interest in the monograph.

I desire also to express my thanks to the Messrs. Wiley and Sons for their promptly executed and painstaking labor in publishing the work.

ADOLPHE MINET.

PARIS, January, 1905.

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PRODUCTION OF ALUMINUM.

PART I.

PROCESSES FOR THE PRODUCTION OF ALUMINUM.

ALUMINUM is found in nature as oxide (Al_2O_3) in corundum, sapphire, and emery; as hydroxide ($Al_2(OH)_6$) in bauxite, hydrargillite, and diaspore; in the form of salts in cryolite (an aluminum-sodium double fluoride with the composition $Al_2F_6.6NaF$), in alum, in the felspars, in slate, and in clay.

The number of processes devised up to the present time for the production of aluminum is very large, but only a few have attained, in their application, an industrial significance. They may be divided into two great and distinctly separate classes:

(A) Chemical Methods. To this class belong the processes devised by Wöhler, Henry Sainte-Claire Deville, Castner, Netto, Grabau, Webster, Frismuth, etc.

(B) Electrochemical Methods. These may be divided into two groups:

(*a*) Electrothermic Processes (Cowles, Héroult, Brin, Bessemer, Stefanite, Moissan with his aluminum-carbide).

(*b*) Electrolytic Processes; namely, those devised by Héroult, Adolphe Minet, Hall, Hampes, Kleiner, Gooch, and Waldo.

A. Chemical Method of Producing Aluminum.

Aluminum was isolated for the first time in the year 1827 by Wöhler,* who produced it impure and in small quantities by means of the effect of potassium upon anhydrous aluminum chloride.

All attempts previously made by Davy, Berzelius, and Oerstedt to decompose the clay by means of the electric current had not yielded the result which, since the successful electrolytic dissociation of the alkali hydroxides, might confidently be expected.

Oerstedt† had attempted, shortly after his discovery of aluminum chloride, to reduce this substance by means of alkali metals, which he allowed to act in the form of amalgams. This was likewise without result.

This noteworthy method, which is the first example of a reduction between anhydrous bodies

* Poggendorffs Annalen, XI, 1827, and Liebigs Annalen, LIII.

† Overs. o. d. Danske Vidensk. Selsk. Forhandl. 1824–1825.

fused by melting, was to be successful for the first time in the hands of Wöhler, who, as we know, isolated beryllium and zirconium as well as aluminum.

The aluminum Wöhler obtained in the year 1827 consisted of a whitish-gray powder having all the physical characteristics of the metals; not until the year 1845 did he succeed in obtaining aluminum in the form of ductile pellets, so that from these he could determine the most important physical and chemical characteristics of aluminum. It was still, however, far from possible to consider aluminum as one of the common metals.

This consummation, the result of a comprehensive investigation of aluminum, which was now about to be obtained for the first time in a perfectly pure condition, was reserved for Henry Sainte-Claire Deville * (1854).

To Deville, furthermore, we owe the first attempt to produce aluminum by the use of metallic sodium; a method whose principle is that of a great number of later patents.

a. Processes based on the Reduction by means of Sodium.

Henri Sainte-Claire-Deville Process.—The first successful improvement made by Deville consisted in the replacement of potassium by sodium; hitherto, following the example of Wöhler, potas-

* Annales de chimie et physique, XLIX, 1854, and St. Claire Deville, *de L'Aluminium*, Paris, 1855.

sium had invariably been employed as a reducing-agent. For the aluminum salt, Deville adhered to the aluminum chloride usually employed.

Thanks to the labors of Brünner, Mitscherlich, Donny, and Mareska in the production of potassium and sodium, Deville was enabled to make for himself without difficulty considerable quantities of sodium, and at the same time to reduce large amounts of aluminum chloride. To this fact, in the main, the final result of his investigations was due.

In the mean time still other obstacles were encountered in the industrial production of aluminum. Apart from the fact that it was necessary to produce great quantities of sodium quickly and cheaply, two other industries must spring into being hand in hand with the manufacture of aluminum: the production and the refining of alumina, and the conversion of this aluminum oxide into anhydrous chloride; to these two processes was added the reduction of the chloride by means of an alkali-metal.

We must not omit to add that, at the time of the above-mentioned investigations of Deville, a mineral rich in aluminum—cryolite (aluminum-sodium double fluoride)—was discovered in Greenland. Deville employed this salt as a flux, adding it in various proportions to the anhydrous aluminum chloride, and found that the chemical reaction took place more readily in the presence of cryolite.

The researches of Deville were begun in the Sorbonne in the year 1854. The first *industrial* investigations were carried out in the establishment of Rousseau in "la Glacière," and were later continued in Nanterre under Morris's direction.

Up to the present time, Deville's process was employed in Salindres, where, on the average, 2000 kg. aluminum was produced yearly. The selling-price, however, was rarely below 100 francs per kilogram.

Rose Process.—In the year 1856 the brothers Tessier established a factory in Amfreville near Rouen, in which aluminum was produced by a process which had been recommended by Rose,* and which depended exclusively upon the reduction by cryolite, a method which had been discovered by Dr. Percy in the year 1855,—a year later, then, than Deville's investigations. We should add, as a matter of historic interest, that before the erection of their factory the brothers Tessier had studied the question of producing aluminum in H. St.-C. Deville's own laboratory.

Castner and Netto Processes. — These processes rest upon the same principle as that of Deville; from an industrial point of view, however, they show a considerable improvement upon the latter, since the price of aluminum produced according to the new method fell below 20 francs per kilogram.

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* Poggendorffs Annalen, XCVI, 1855.

Castner Process.*—This process, which was employed about 1899 by the Aluminum Company, Limited, in Oldbury, Birmingham, shows substantial improvements, mainly in two directions: (1) in the production of the sodium with the aid of caustic hydrate of soda at a low temperature, and (2) in the production of aluminum-sodium double chloride.

Production of Sodium.—Castner obtains sodium from caustic hydrate of soda with the aid of an artificial iron carbide at a temperature which does not exceed 1000° C., and which is, therefore, considerably lower than the temperature reached by Deville. The latter availed himself in this case of the classic method of reduction, with soda, by means of carbon.

The composition of the iron carbide employed is expressed in the formula FeC_2; from 7 to 8 kg carbide were mixed with 12 kg caustic hydrate of soda, and gave as a product of reaction 2 kg sodium, according to the equation

$$6NaOH + FeC_2 = 6Na + Fe + CO + CO_2 + 3H_2O.$$

The iron in this process, then, merely plays the part of an intermediary substance.

Production of Aluminum-sodium Double Chloride.—The difficulty in this process is to secure a steady supply of chlorine gas. The chlorine, generated

* Cf. *L'Aluminium, fabrication et emploi*, by Adolphe Minet, pp. 127–137.

by the Weldon process, is first collected in lead gasometers, and then conducted over a mixture of clay, carbon, and common salt, which is contained in a horizontal retort 3.6 m in length; the retort is heated by means of gasoline gas. The mixture containing aluminum is dephlegmated in the apparatus even before it is treated with chlorine, and the resulting aluminum-sodium chloride, according to the form and proportion it takes, is condensed in brick receptacles.

However pure the material from which it is derived may be, the chloride produced in the method described will still be found to include, invariably, considerable quantities of iron; and since the weight of the chloride should be about ten times as great as the weight of the aluminum produced, it follows that the metal obtained in this manner would contain far too large a proportion of iron, were not the double chloride, before it is decomposed by sodium, subjected to a special purification. This consists in melting it with a small quantity of aluminum- and sodium-powder. The proportion of iron, which in many cases originally amounts to 1%, is decreased by this treatment to 0.1%.

Another method for purifying the double chloride depends upon its treatment electrolytically: a method likewise proposed by Castner.

Reduction of the Aluminum Chloride. — The chloride is mixed with cryolite in the proportion

2:1, with the addition of small pieces of sodium; the whole is then mixed in a rotating cylinder, which is introduced into an air-furnace previously heated to the temperature of reaction. The charge consists usually of 550 kg double chloride, 150 kg cryolite, and 150 kg sodium. The quantity of aluminum obtained by means of a single operation amounts to about 60 kg.

Netto Process.* — This process was operated by the Alliance Aluminium Company in Wallsend at Newcastle, and, indeed, simultaneously with Castner's process. Netto's method is a modification of the old cryolite process, as it was first proposed by Deville, and industrially introduced by Rose and Percy in the year 1885. It depends upon the reduction of cryolite by sodium, and may be divided into three important parts: 1. The production of sodium. 2. The production of cryolite. 3. The treatment of the cryolite with sodium.

Production of Sodium.—Netto obtains the sodium professedly in a very economical fashion, by allowing glowing coke to act upon caustic hydrate of soda. His apparatus (Fig. 1) consists of a cast-iron retort, *b*, which is filled with coke and charcoal and brought to a red glow. In the upper part, by means of the mouthpiece *d*, the caustic hydrate of soda is introduced, which is melted in the receptacle *e*. While the caustic hydrate is falling drop by drop upon the

* Cf. A. Minet, *L'Aluminium*, pp. 132–137.

glowing charcoal, it dissolves almost instantaneously. In the condenser *g* the sodium-vapor generated is condensed.

To produce 100 kg sodium are needed: 1000 kg NaOH, 120 kg casting-pieces, 1200 kg fuel, reckoned as coke, and 150 kg charcoal as reducing-agent.

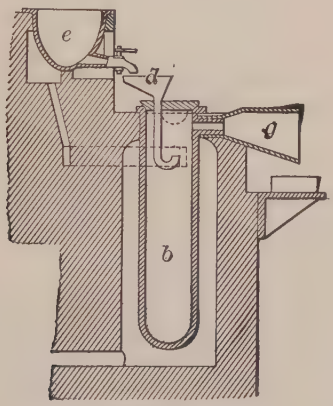

FIG. 1.

Production of Cryolite.—Netto uses for the production of this substance the slag which results from the treatment of the cryolite with sodium, and which essentially consists of sodium fluoride.

If one mixes sodium fluoride with aluminum sulphate, and heats the mixture to the melting-point, there results the following reaction:

$$12NaF + Al_2(SO_4)_3 = Al_2F_6.6NaF + 3Na_2SO_4;$$

the sodium sulphate, which is formed simultaneously with the cryolite, is separated from the latter, after

having been previously cooled, merely by lixiviation.

Treatment of Cryolite with Sodium—For the success of this process it is one of the most essential conditions to have the alkali-metal affect the cryolite as quickly as possible, in order to avoid the excessive loss of sodium by vaporization, and to prevent too strong an attack upon the fire-bricks and upon the natural and manufactured fluorides always found in association with silicates. By means of a number of very ingenious contrivances Netto actually succeeded in materially shortening the time of the reaction. Each charge gives a product of about 5 kg aluminum.

Grabau Process.* — In order to prevent the troublesome consumption of the fluoride, Grabau treats sodium and aluminum fluoride separately (producing the latter himself), and allows the substances to react on each other, the aluminum powdered, the sodium in the form of small cubes or cylinders. The result, with the simultaneous development of a considerable amount of heat, is the following reaction:

$$2Al_2F_6 + 3Na_2 = Al_2 + Al_2F_6.6NaF.$$

After the mass has been cooled down, the aluminum is found as a regulus on the bottom of the

* D. R. P. (Ger. Pat.) 47031.

crucible, covered with a slag of cryolite, which during the reaction is melted.

Production of Aluminum Fluoride. — Aluminum sulphate and cryolite are mixed in equivalent proportions; heated, the following reaction takes place:

$$Al_2(SO_4)_3 + Al_2F_6.6NaF = 2Al_2F_6 + 3Na_2SO_4.$$

The aluminum fluoride, since it is insoluble in water, is separated from sodium sulphate by filtration.

Production of Sodium.—Grabau obtains sodium by the electrolysis of molten sodium chloride. The principal original feature in his process is the form of the electrolytic apparatus (Fig. 2).

The double-walled porcelain receiver *BB* forms the significant feature of the apparatus; this receiver encloses the negative iron electrode *n*. The current enters through the carbon anodes *CC*, flows through the electrolytes (melted chloride of sodium) both without and within the polar cell *BB*, and makes its exit at *n*. The chlorine passes out at *d*. The sodium, since it is lighter than its melted chloride, mounts up within the receiver and escapes through the tube *a*, whence it is conducted into the condenser *M*, which, filled with nitrogen or hydrogen, is sunk in a reservoir *S* containing petroleum. The screw *H* serves to remove clogging material which may eventually collect in the tube-

shaped portion E of the negative electrode. In addition to the above-mentioned process, there are still other well-known processes * for obtaining sodium electrolytically, such as those of Castner

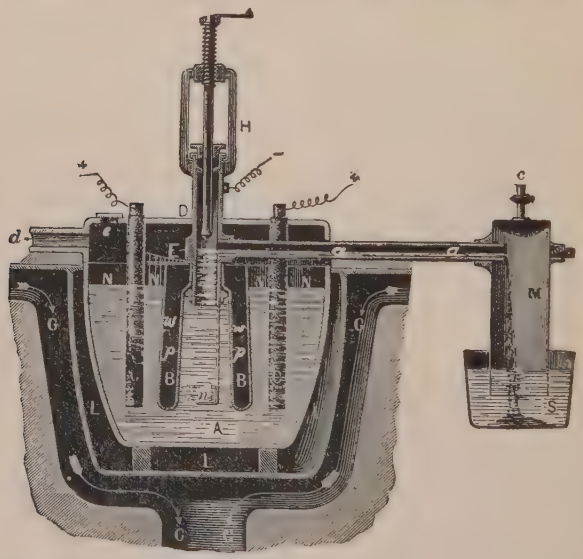

FIG. 2.

(1890), Minet (1890), Borchers † (1893), Becker (1900).

Frismuth Process.—Aluminum-sodium double chloride is volatilized in the retort, in a chlorine current, in the presence of common salt; upon the chloride, in the form of vapor, sodium-vapor in a

* Cf. A. Minet, Traité théorique et pratique d'électrometallurgie, p. 415 ff.

† Borchers, Alkalimetalle, in Zeitschrift für angewandte Chemie, 1893.

suitable receptacle is allowed to act; this sodium-vapor is formed in a peculiar retort from a mixture of soda and carbon heated to a red glow.

Webster Process. — This is based upon the same principle as the process of Deville. The original material is alum, from which the aluminum chloride is produced. By this means the two principal impurities of aluminum, namely, percentages of iron and of silicon, are avoided.

The Webster process was utilized by the Aluminium Crown Metal Company in Holyhead, at Birmingham.

The White and Thompson Process.—This operates similarly to that of Rose. Three parts of sodium and four parts of powdered cryolite heated to 100° C. are mixed in a sand-bath, thoroughly stirred and allowed to cool. To this are added four parts of aluminum chloride, and the whole is then placed in an air-furnace, heated to a red glow, whereupon the reduction begins immediately.

Feldmann Process (Linden vor Hannover).— A mixture of aluminum-strontium double fluoride, strontium chloride, and sodium is heated to the melting-point, whereupon the following reaction occurs:

$$Al_2F_6.3SrF_2 + 3SrCl_2 + 6Na =$$
$$2Al + 3SrF_2 + 3SrCl_2 + 6NaF.$$

The strontium fluoride, since it is insoluble in water, can be separated by washing it away from

the other constituents; and thus, returning into the process, it serves for the production of additional quantities of the double fluoride.

b. **Processes which do not employ Sodium.**

Under this heading we shall speak of those methods which effect the reduction of aluminum without the aid of an alkali-metal.

Apart from the Beketoff process (1865)—Beketoff proposes magnesium as a means of reduction—we may here mention:

Reillon, Montagne, and Bougerel Process.— This depends on a reaction the correctness of which has not been demonstrated: the production of aluminum by heating a mixture of clay, carbon, and bisulphide of carbon, upon which a hydrocarbon is allowed to react.

Baldwin Process (Chicago).—This is based on an insufficiently defined reaction, which in my opinion cannot be verified: bauxite, powdered carbon, and common salt are so to react upon one another under the influence of heat that an aluminum-sodium compound is formed. The alloy obtained is furthermore to be melted with a quantity of sodium chloride, and thus the aluminum is to be separated from the alkali-metal.

Faurie Process.—A quantity of sulphur, carbon, and clay is heated to a red glow. First, aluminum sulphide and bisulphide of carbon will be formed, and finally, at a white heat, aluminum.

Stephen and Sanderson Process.—The details of this process are not exactly known. On the one hand, fluorhydric acids in a gaseous state are allowed to act upon a quantity of alum and emery heated to a red glow until the whole mass becomes of a pasty consistency. From the melt, grains of aluminum containing iron are deposited, which may be purified from the iron by dilute sulphuric acid. If it is desired to obtain an alloy of aluminum and iron, hematite is added to the melt.

On the other hand, zinc may also be used as a reducing-agent, which, acting in the form of vapor overshot above white-hot aluminum chloride, is said to reduce the latter. A residuum will be formed at the same time, containing zinc as an impurity; by heating to 1100° C. this zinc may be gotten rid of.

Pearson and Pratt Process.—According to the proposal of these engineers, iron-aluminum alloys are obtained in blast- or cupola-furnaces directly from aluminum ores. The latter, with this end in view, are mixed with iron ores as rich as possible in clay, and with calcium fluoride (fluor-spar)—instead of common lime—whereupon the mass is introduced into the blast-furnace.

If it is desired to produce aluminum steel, the original materials must be free from sulphur and phosphorus. The melt, containing aluminum, is then handled by the Bessemer process in the usual manner.

The place of the lime may to advantage be wholly taken by fluor-spar; with a substitution up

to 25%, satisfactory results are nevertheless achieved. Thus, for example, in the case of the ores of Staffordshire, which are especially rich in protoxide of iron and alumina, a charge has the following composition: 40 parts clayey ores, 11 parts lime, 4 parts fluor-spar, and 60 parts carbon with a blast of hot air; an equal amount of coke with the blast of cold air.

Stéfanite Process.—This patent is quite similar to the one just described, and has been employed particularly in Germany. It consists essentially in adding to the usual blast-furnace charge emery or alum in powder or briquette form. A melt containing aluminum is thus obtained, which, under further treatment in the puddling-furnace, gives a metal which permits of being hardened like steel and, according to the statement made in the description of the patent, is much more capable of resistance than iron.

Other Processes.—For the sake of completeness we add still other processes in the following table, although these have been practically tested in but few instances.

Process.	Proposed Means of Reduction.
Knowles and Corbelli	Cyanogen gas
Gerhard and Fleury	Hydrocarbon
Morris and Chapelle	Carbon
Morris	Carbonic acid
Lautherborn and Nieverth	Iron
Calvet and Johnson Beuson	Copper
Dulls, Basset, and Seymour	Zinc
Wilde	Lead
Weldon	Manganese

The original material in the case of all these attempts was either the oxide or the chloride or fluoride of aluminum. It must be emphasized that all these reductions were attempted without the assistance of electricity. If, therefore, there has been no practical result from these processes up to the present time, as we have already stated, it is still not impossible that by the use of the electrical current in this connection one or the other of these processes may result at least in the formation of alloys of aluminum.

B. Electrochemical Methods of Producing Aluminum.

The processes for the production of aluminum with the aid of the electrical current may be divided, as, indeed, is true of all electrometallurgical methods, into two groups: the electrothermic and the electrotytic; to these may be added those processes in which both functions of the electric current are simultaneously active, and which one may call combined processes.

Electrothermic Processes. — Electrothermic processes are those in which the current plays merely the part of a heating agent, regardless of whether the calories are delivered by an electric arc or by a resistance with a current flowing through.

In the first case the electromotive force is nearly that of the usual electric arc, amounting, therefore,

to something like 30 or 35 volts; in the second case the tension depends upon the circumstances for the time being. The resistance material may now be either independent of the reaction-substances—and then we have to do with a purely electrothermic process—or the reaction-substances are themselves the ones that form the resistance, and then, under certain conditions, the electrothermic process may be accompanied by an electrolytic process (the combined process).

In purely electrothermic processes the direct current may be used as well as the alternating.

The quantity of heat Q developed by the current passing through may be calculated by the formula

$$Q = kEJt \text{ Cal.}$$

E signifies the difference in potential in volts between the point of entrance and the point of exit of the current, whereby only that region comes under observation which by virtue of its slight conductivity produces current heat; J is the strength of current in amperes, t the time, expressed in seconds; k is a factor of proportion, which with regard to the chosen units amounts to the value 0.24. We have, then, the relation

$$Q = 0.24\, EJt \text{ Cal.}$$

The current-density, that is to say, the strength of current per square centimeter, amounts generally to 10 amp.

Electrolytic Processes.—Here the current operates in twofold fashion. On the one hand it develops heat by its passage through the electrolyte, on the other hand it brings about the electrolytic decomposition of the electrolyte. The electrolyte is then found to be in a liquid state, whether as a melt or as a solution. It goes without saying that in this case only the direct current may be employed.

The weight of the matter separated by the electrolysis is proportional to the amount of current, or, at a given time, proportional to the intensity of current J (Faraday's law). The electromotive force, which in the case of soluble anodes amounts to only a few tenths of a volt, whereby the electrolytic process confines itself merely to carrying the element in question from one electrode to the other, in the case of insoluble anodes, and therefore in the case of a decomposition peculiarly electrolytic, rarely exceeds 5–6 volts.

The choice of current-intensity at the cathode is governed by the particular type of electrolysis, by the nature of the metal separated, and also by the temperature. In the case of aqueous solutions the intensity varies between 0.001 and 0.01–0.02 amp., in the case of molten fluxes between 0.5 and 1 amp.

Work of the Current.—This is expressed in the formula

$$Q = 0.24 E J t \text{ Cal.},$$

in which the factors E, J, and t have the same meaning as before.

A part of the energy is converted into heat; let Q_1 be this portion; we then have

$$Q_1 = 0.24 R J^2 t \text{ Cal.}$$

R is the resistance in ohms of the electrolyte.

The remainder Q_2 is equivalent to the energy-expenditure of the chemical process taking place in the electrolysis. Its value is expressed by the formula

$$Q_2 = 0.24 e J t \text{ Cal.},$$

if we characterize by e the counter-electromotive force of the decomposition.

We have then

$$Q = Q_1 + Q_2,$$

or

$$0.24 E J t = 0.24 R J^2 t + 0.24 e J t.$$

If we abbreviate this on both sides, we obtain

$$E = e + R J,$$

an expression which gives us the principal formula for all electrochemical processes.

Combined Process.—Under this heading should be classified all reactions in which an electrothermic phenomenon is accompanied and converted by an electrolytic. Let us suppose, for example, that the arc serves as a source of heat, that the electrodes

are at the beginning of the process independent of the materials of reaction, and that the electromotive force is about 35 volts. As soon as the charge, in consequence of heating, melts, and its volume is increased by filling up, thus coming into contact with the electrodes, the electromotive force may sink to about 20 volts or even lower, and the arc disappears. The electrothermic phenomenon, it is true, still remains predominant, as the heat-effects appearing in the neighborhood of the electrode-surface demonstrate; on the other hand, however, there takes place a more or less well-defined electrolytic process, which in the case of the direct current diminishes, in the case of the alternating current increases the product. Simultaneously the current-density gradually falls; this, in the case of a purely electrothermic process, amounts to 5–10 amp. If we have the direct current, then the phenomena of warmth disappear forthwith, the difference in potential and the current-density approach the values observed universally in the case of salts melted by electrolysis; in brief, the electrolytic effect of the current predominates over the electrothermic, which latter may, indeed, finally disappear altogether.

(*a*) Electrothermic Processes.

Alumina (Al_2O_3) is reduced by means of carbon or a metal, with or without the addition of a flux. The aluminum produced by this means is not

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pure, but forms according to the method of the operation an alloy (Cowles, Héroult) or a carbide (Moissan).

Moukton Process.—The application of the electric current to the reduction of alumina by means of carbon was proposed for the first time by Moukton in the year 1862. According to his patent the electric current is to be conducted through a reduction-chamber charged with carbon and alumina, and the mixture thus brought to the temperature requsite for the reduction. With reason, however, Borchers emphasizes the fact that the process mentioned, even had Moukton been able to produce a metal industrially available, would not have been profitable; for not until long after the invention of the dynamo-electric machine (1872) did it become possible to maintain electrical energy inexpensively. Apart from this, however, Moukton would have been able, according to his patent, to produce merely a totally useless aluminum-carbide, and by no means a metal satisfying the demands of industry.

Cowles Process.—Only after a considerable number of years, during which the application of electricity to metallurgy seemed to have passed entirely into oblivion, did the Cowles brothers (1884) come forward with a process which yielded, if not pure aluminum, at least alloys containing aluminum up to 20%.

The characteristic feature of this Cowles inven-

tion is the utilization of a type of apparatus which is styled the electric furnace, and which is rightly considered to be the first great advance in electrometallurgy.

Electric Furnaces. — It is true that there were already before Cowles apparatus which might be enumerated in the group of electrical furnaces; still, they were not all capable of being utilized for technical purposes. Such were the furnaces of Depretz * (1849), Johnson † (1853), and Pichon ‡ (1853). And furthermore the operations of Berthelot (1862), Siemens § (1879), and Louis Clerc ‖ (1880) did not advance beyond the limits of laboratory experiments.

Although the Cowles brothers constructed for the first time a practical furnace, in which considerable quantities of electrical energy were converted, the credit of having anticipated the now perfected apparatus and a great number of more recent constructions of various forms belongs to the chemist Héroult and to those who showed how to produce *pure* aluminum.

Cowles Furnaces. — Eugene and Alfred Cowles built several furnaces for specifically electrothermic purposes, that is to say, they were furnaces in which

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* Compt. rend. de l'académie des sciences, Dec. 17, 1849.
† Engl. Pat. No. 700 of 1853.
‡ According to Andreoli, Industrie, 1893.
§ Engl. Pat. No. 2110 of 1879.
‖ Elektrotechn. Zeitschr., 1880.

the reduction of metallic oxides by chemical means with the coöperation of the current heat was carried out.

First Type.—This (Fig. 3) is the subject of a patent of the year 1885.

The material with which the furnace is to be charged is introduced in minute form, mixed with retort-carbon in fine grains, and is heated to a white glow by the heat developed by the passage of the

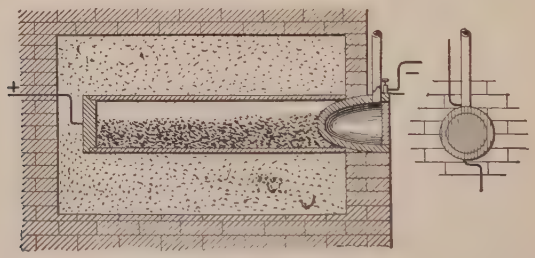

FIG. 3.

current. The furnace was originally designed for the reduction of zinc ores, but was used eventually for other ores also, in particular for obtaining aluminum, magnesium, boron, etc. The furnace is built with a retort of cylindrical form, which is made of silica or some other current-insulating material. It is surrounded by granulated charcoal or some other poor conductor of heat, and is shut off on the one end by a plate of carbon, which serves as positive electrode, on the other end by a graphite crucible, which provides the negative

electrode. The latter was originally also designed to be a condensing-chamber for the zinc vapors.

Second Type (patented in the year 1886).—This furnace (Fig. 4), which is based on the same principle as the previous one, has the form of a parallel-

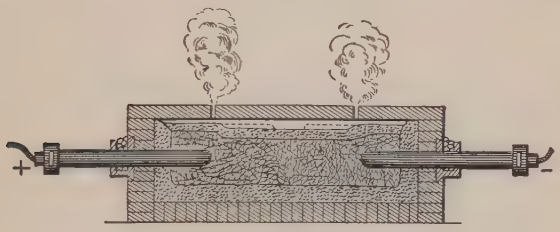

FIG. 4.

opipedal chest of masonry. The two bar-shaped carbon electrodes are introduced directly into the reaction-mixture, which rests upon a support of insulating materials. At the beginning of the operation the electrodes are made to approach one another until there is an opposite contact; the portions of the furnace-charge that are in proximity to the ends of the electrodes forthwith come to a white heat, whereupon the carbons are again slowly separated from one another to the distance seen in the design. After the closing of the current, immediately after the separation of the electrodes, the distance between the opposite electrodes amounts to about 25 mm; the distance increases to 1.2 m at the close of the operation.

Third Type.—This furnace, which, like the preceding, was patented in 1886, has stood the test of

practical experience best of all the Cowles apparatus; it has had a very wide-spread employment in technology.

The furnace (Figs. 5 and 6), whose walls are

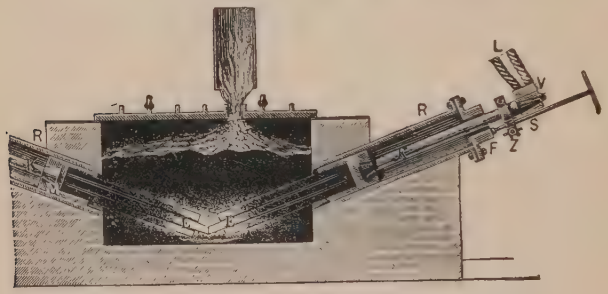

FIG. 5.

constructed of fire-brick, has an average height of .66 m; the broad side is 1.68 m, the narrow side 0.51 m. At the lower edge of one of the sides is

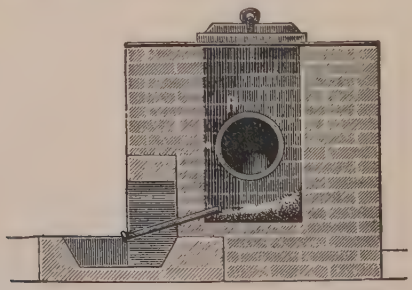

FIG. 6.

an opening for emptying out the contents of the furnace (comp. Fig. 6). In both side walls two cast-iron tubes are set in, which make an extension of the carbon bars that serve as electrodes.

Each electrode consists of nine such carbon bars about 6 cm in diameter and 80–97 cm in length. They are of iron or copper rod, according to the alloy which is to be produced; the rod is provided with a female screw, in which a screw is inserted, by means of which the electrodes may be thrust into the cast-iron tube.

The first technical tests with this furnace were carried out in Cleveland with an available pressure of 50 volts and a current-strength of 1500 amp. The power (100 electric horse-power) was furnished by a steam-engine.

The first thing was to produce pure aluminum. With this end in view, the reduction of the alumina by means of carbon was attempted; but these efforts were unsuccessful. Conditions became more hopeful when the place of carbon was taken by a metal, iron or copper, and so, without difficulty, the iron and copper alloys of aluminum were obtained.

Later the Cowles brothers erected a factory in Lockport, where there was water-power at their disposal able to furnish a stream of 3000 amp. and 50 volts (200 horse-power). In Milton, also, a factory was started, which carried on the industry by the method described, and made use of a 400-horse-power dynamo-machine for generators, the machine at 60 volts pressure giving a current of 500 amp.

As material for producing ferro-aluminum a

mixture of bauxite, iron filings, and powdered carbon may be used. For the production of copper-aluminum, bauxite, since it is too rich in iron, is unsuitable. Emery or corundum is used. In addition, it is needless to say, copper metal takes the place of the iron rod. The proportion of aluminum varies between 5 and 20% in ferro-aluminum, and between 18 and 30% in copper-aluminum.

The costs of the electrical energy per kilogram of finished aluminum were in Lockport equivalent to the price of 77 horse-power hours; that is to say, a horse-power hour gave 13 g pure aluminum or 65 g 20% alloy. In Milton, on the other hand, in round numbers 40 horse-power hours were necessary for the production of a kilogram of aluminum; this means a production of 25 g pure aluminum or 125 g 20% alloy per horse-power hour.

Fourth Type (Fig. 7).—This possesses a great similarity to the furnace of Johnson of the year 1853; we mention it here for the sake of completeness. The details may be seen from Fig. 7.

Héroult Processes.—The experiments of Héroult, which date from the year 1886 and which are not yet concluded, concern themselves with almost every department of electrometallurgy: the electrothermic production of aluminum alloys, of silicon and its alloys, of calcium carbide, of steel, etc., and furthermore the production of pure aluminum electrolytically.

Of these various processes, which are described

in a great number of patents,* we shall single out for especial mention those which operate electrothermically; and of the types of furnace constructed by Héroult we shall mention only the more important.

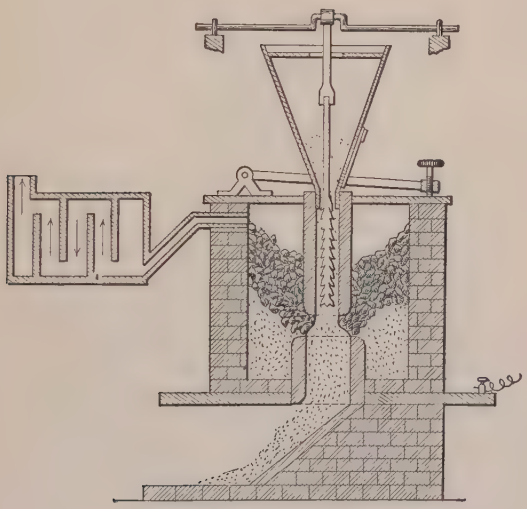

FIG. 7.

During the first ten years of his investigations Héroult constructed practically but one type of furnace — that which the French to-day style "cuve-cathode" (crucible-cathode). After manifold modifications in the design originally conceived,

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* French pat. No. 170,003, April 15, 1887.
Belgian " " 77,100, " 16, "
English " " 7,426, May 21, "
German " " 4,165, Dec. 8, "
American " " 387,876, Aug. 14, "

and after repeated improvements, he finally brought his furnace to such a state of perfection that its technical employment is at present very wide-spread.

First Type of Furnace (Fig. 8).—It reminds one

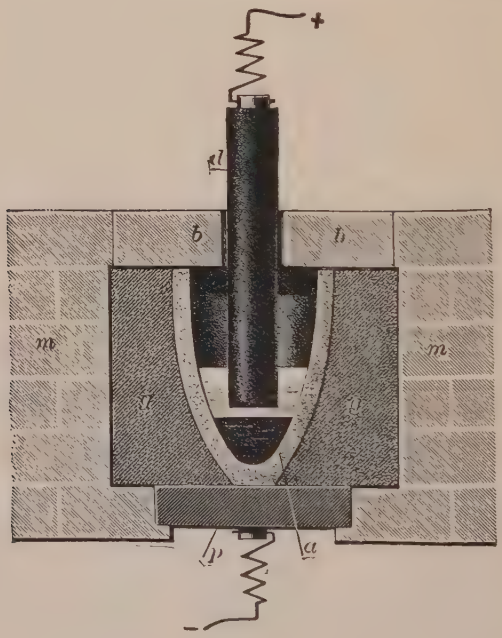

Fig. 8.

of the furnace-construction of Siemens, which is represented in Fig. 9.

We must, however, lay stress upon the fact that the Héroult furnace from a technical point of view marks a significant advance; it is built more strongly than the Siemens furnace, and while the

German investigator employed his furnace merely for laboratory experiments within narrow limits, namely, for the electric smelting of metals, his apparatus served the French chemist in the years

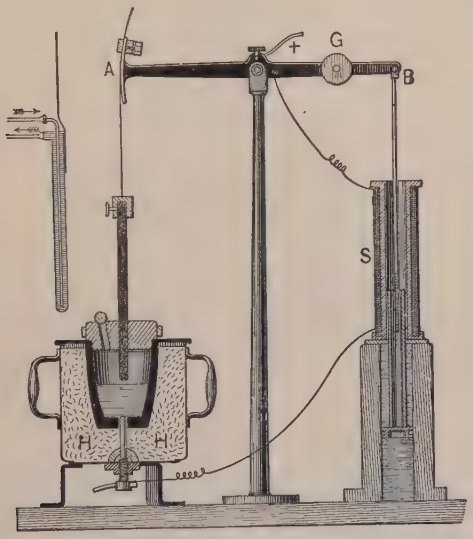

FIG. 9.

1886 and 1887 for obtaining aluminum alloys, in particular aluminum bronze.

In his patents Héroult characterizes his invention as "a process for the production of aluminum alloys with the aid of the thermic and electrolytic effect of the electric current upon aluminum oxide (Al_2O_3) and the metal with which the aluminum is to be alloyed."

In order to keep the reduction-material more

easily in a molten state, and so accelerate the reduction, Héroult adds also a few parts of cryolite.

Second Type of Furnace.—For the first experi-

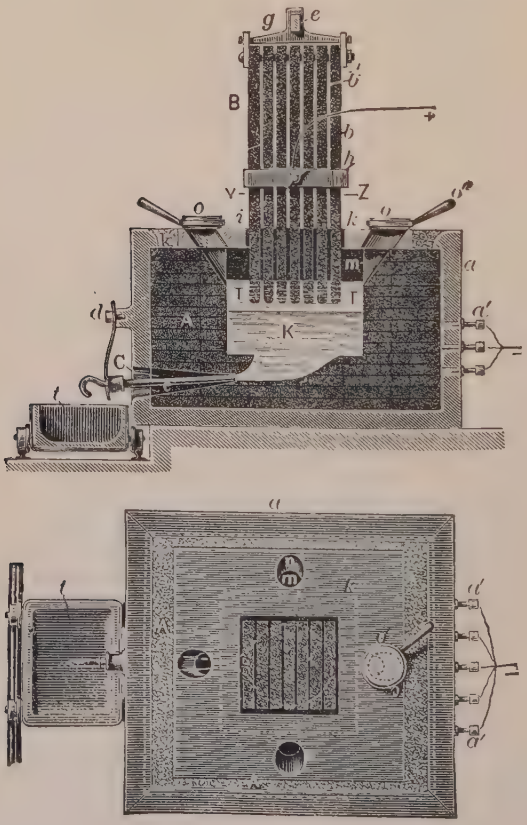

FIG. 10.

ment, which was carried out on a small scale only, the furnace just described answered all purposes, although it was necessary after each smelting to

take out the crucible in the middle of the furnace in order to empty it, a rather troublesome operation, and one necessarily entailing a loss of time.

This disadvantage was obviated by a second form of construction (Fig. 10), in which the finished product at definite time-intervals may be set free through a tapping-hole *C*.

The Schweizerische Metallurgische Gesellschaft, later the Aluminium-Industrie-Aktiengesellschaft, at Neuhausen, Switzerland, undertook to operate this furnace, which Héroult had described in his first patent. It required a current of from 12 to 15 volts and 13,000 amp. (200–250 horse-power), and yielded per horse-power hour 25–30 g aluminum, which was obtained in the form of a copper alloy containing 15–20 % of aluminum.

Third Type of Furnace.—In the year 1890 certain technical journals* published the description of a new Héroult furnace, which in Neuhausen and in Froges, France, was employed especially for the electrolytic production of aluminum. From Fig. 11 it may be seen that this furnace is merely a modification of the preceding.

Fourth Type of Furnace.—This was constructed by Héroult in collaboration with Kiliani,† likewise for obtaining pure aluminum electrolytically. It is represented in Fig. 12.

Its characteristic feature consists in an arrange-

* Industries, VIII, 1890, p. 499.

† D. R. P. No. 50508, April 21, 1889.

ment which makes possible a continuous rotation of the positive electrode, and which clearly subserves the purpose of retarding the phenomena of heat such as are often observed during the elec-

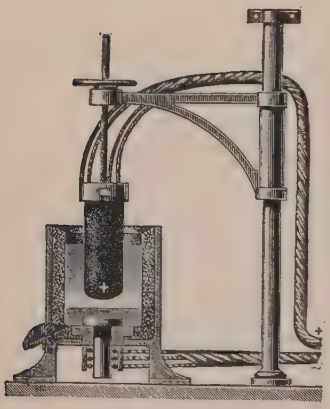

FIG. 11.

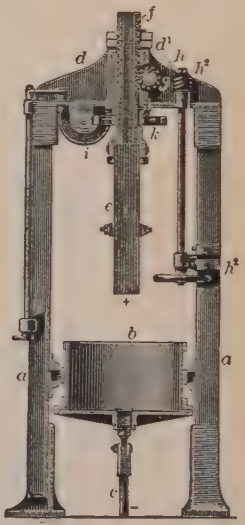

FIG. 12.

trolysis of salts at the fusing-point. We shall see that these phenomena occur only beyond a certain current-density, and that they become fairly rare when one is working with several apparatus regulated for pressure.

Borchers Furnaces. — Borchers has built a large number of furnaces, some for the reduction of oxides and the electrothermic production of metal carbides, and some for the electrolytic production and refining of certain metals. Since most of them

may find a use in the electrometallurgy of aluminum, a description of them is given.

Electric Furnaces for the Production of the Carbides of the Earth-alkalies. — In the years 1880–89 Borchers succeeded, by means of carbon at a high

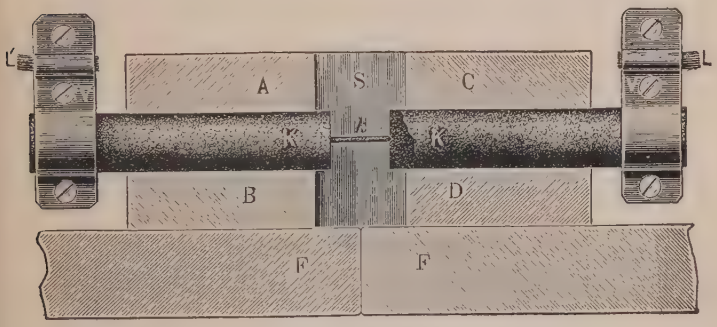

Fig. 13.

temperature, electrically developed, in reducing all the metallic oxides till then regarded as unreducible; since, however, he always worked with an excess of carbon, he could only produce carbides

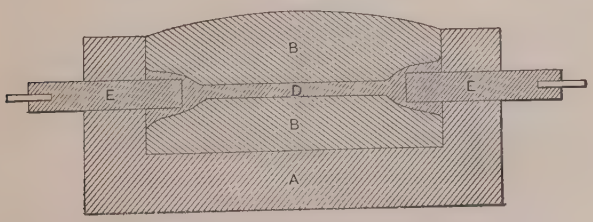

Fig. 14.

which contained free carbon. Not until twelve years later did the French chemists Moissan and

Bullier succeed in obtaining well-defined carbides, which were free from carbon in excess.

First Type.—For his first experiments Borchers employed an apparatus for currents of 12 volts and 120 amp. (2 electric horse-power). Its details may be seen in Fig. 13. It may readily be constructed with fire-brick and bars of carbon.

Between two carbon bars *KK* 40 mm in diameter, a thinner rod of carbon *k* only 4 mm in diameter and 40 mm long is fastened. Through a suitable arrangement of fire-brick round about *k* a cavity is left free, which is filled with a mixture of oxide and carbon. Within even a few minutes after closing the circuit the whole mass between the carbon bars *KK* is turned into carbide.

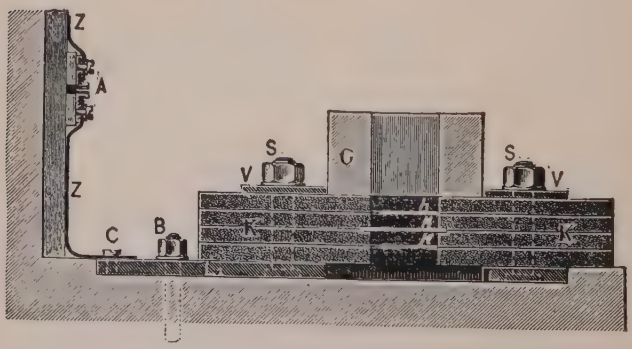

FIG. 15.

A furnace quite similar (Fig. 14) was employed in 1890 by Acheson for producing corundum (silicon carbide).

Second Type.—The arrangement just described is also practicable, with certain modifications, upon a greater scale. Figs. 15 and 16 represent the longitudinal and the cross-section of a furnace built

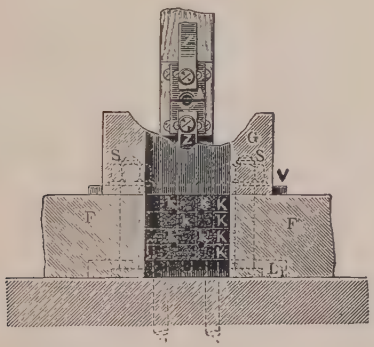

FIG. 16.

for currents of 24 volts and 610 amp. (20 electric horse-power).

The massive carbon bars of the laboratory furnace, Fig. 13, are here replaced by carbon plates *KK*, between which three small carbon bars *kkk*, 4 mm in diameter and 80 mm in length, are introduced.

Electric Furnaces for the Reduction of Metallic Oxides by means of Carbon. — For this purpose likewise, having in view the production of pure metals or alloys, Borchers has planned certain furnaces, the principal types of which we will now describe.

First Type.—This finds a use also in the electrometallurgy of aluminum. Between two large car-

bon bars *KK* (Fig. 17), of 25-30 mm diameter, a thin carbon rod *W*, about 3 mm in diameter and

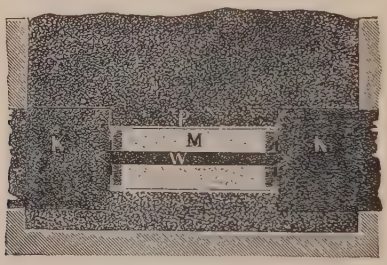

FIG. 17.

45 mm long, is fastened. This lies in the axis of a small paper cartridge, about 40 mm in length, which is filled with a mixture of clay and carbon. After the cartridge has been covered over with coarse carbon powder the circuit is closed (current about 35–40 amp.). The reduction is complete at the expiration of three to four minutes. After cooling down, the carbon rod *W* is found to be surrounded by a mass which consists of aluminum rich in carbon.

If copper or copper oxide is added to the furnace-charge, one obtains, instead of a metal containing more or less carbon, a copper-aluminum alloy.

The current-strength specified expresses a current-density of 500 600 amp. per square centimetre of cross-section, measured at the middle copper rod *W*; if the current-density be increased to 1000 amp.,

it is possible to melt with the apparatus even the most refractory metals.

The pressure required for a current-density of 500–600 amp. amounts to 10–17 volts.

Chronologically, these investigations of Borchers—and this we are not willing to pass over without mention—are three or four years later than the experiments, already described, of Cowles and Héroult.

Second Type.—Fig. 18 shows one of the simplest forms of the Borchers furnace. The graphite crucible *T* contains the mixture *O* of oxide and carbon which is to be reduced, and represents at the same time one of the two electrodes, whilst the other is formed by the massive carbon bar *K*. Between the two the thin carbon rod *W* is introduced. *S* is a fire-brick covering for the crucible.

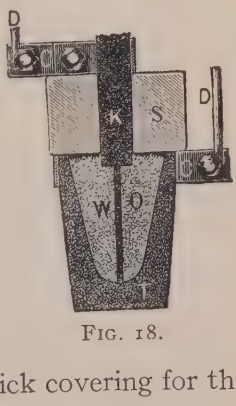

FIG. 18.

Third Type.—Two further forms of technical apparatus * are represented in the three following illustrations. One (Figs. 19 and 20) rests on a plate *F*, which is provided with two backs, of which the one (*B*) is fastened to the foot-plate, or is cast in the same piece with it, while the other

* Borchers, Proben, in Zeitschrift für angewandte Chemie. 1892, p. 133

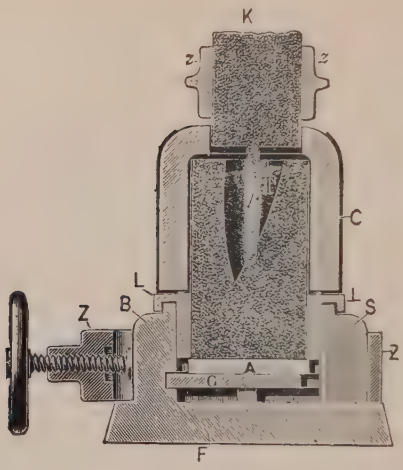

FIG. 19.

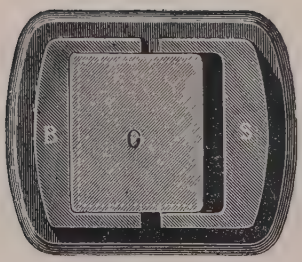

Fig. 20.

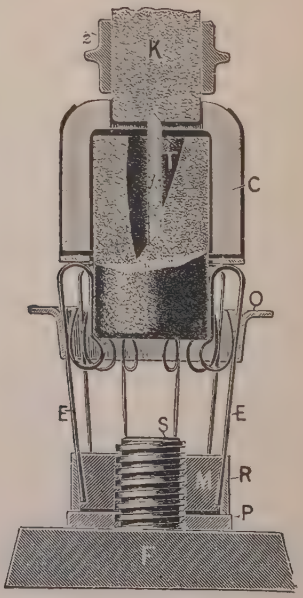

FIG. 21.

(*S*), through the iron band *Z*, by means of a screw or spring, may be made to approach the back *B*. To both backs a contrivance is attached for the purpose of receiving the iron plate *G*. This arrangement serves on the one hand to support the crucible *T*, on the other hand to guide securely the back *S* as it approaches *B*.

If necessary, that is to say, if the crucible is no longer sufficiently high, a fire-brick or asbestos plate is placed beneath. The remaining portions of the apparatus, as well as the fire-brick covering *c*, the carbon bars *K* and *k*, the carbon-container *z*, are similar to the corresponding parts of the furnace last described.

Another and a very simple form of crucible-holder is seen in Fig. 21. It serves to hold firmly crucibles of various diameters.

Upon an essentially different principle, namely, the electrolysis of melted aluminum compounds, is based a furnace which is represented in Fig. 22, and which was constructed by Borchers especially for the electrometallurgical production of aluminum.

T is a crucible with fire-brick bottom *B*, the interior of which is entirely lined with a mantle *F* of alumina or some other refractory aluminum compound. In the floor-lining a steel plate *K* is inlaid, into which the copper tube *R* is screwed; this tube may be cooled by water or by some other suitable means. The cold water is introduced through a narrow tube *E*, while the warm escapes

through the tube X, which reaches almost to the upper end of the copper tube R. The latter, through the rivet V and the cable N, is connected

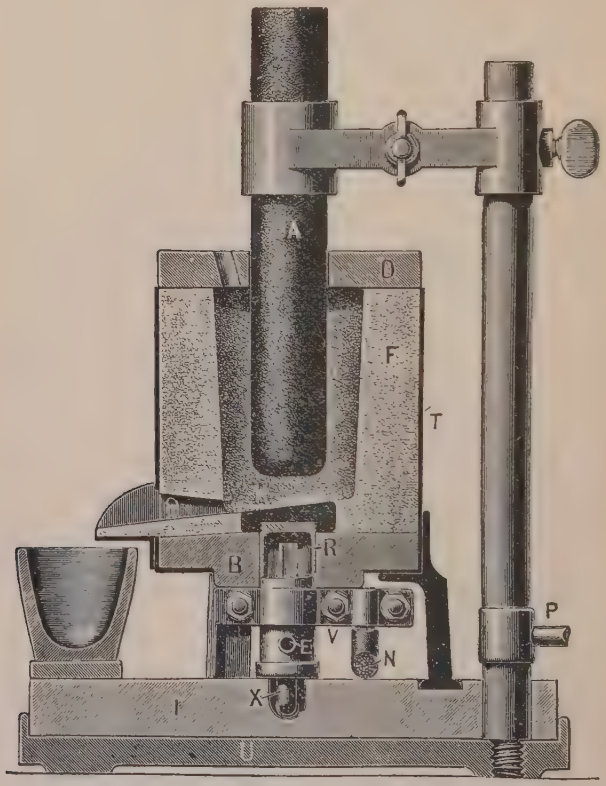

FIG. 22.

with the generator, and is thus the means of conducting the current to the steel plate K, which at the beginning of the operation serves as cathode. The anode is the massive carbon bar.

Apparatus of Borchers' construction were built by the firm of E. Leybold's Successor in Cologne, in a form convenient for experiments, for currents of 120–200 amp. and 5–12 volts.

Of the furnace types mentioned, the last has been tested with especial thoroughness; we should, however, remark that the Héroult and the Minet crucible-cathode furnaces preceded it, which, moreover, operate with currents of 6000 amp. and 8 volts (65 electric horse-power), when used in the electrometallurgy of aluminum, and with a like strength of current require 32 volts (260 electric horse-power) when used for the production of calcium carbide.

Willson Process.*—The first furnace which was constructed by the American engineer Willson for the electric preparation of aluminum compounds (Fig. 23) reminds one of the apparatus of Héroult and Borchers. His process is based on the reduction of alumina by the electric arc. Later Willson devised still another arrangement (Fig. 24) which originated in the idea of saving as much as possible the wear and tear upon the anodes in the electric furnaces which operate according to the combined electrothermic-electrolytic process. To this end he gave the carbon anode the form of a

* American pat. No. 430453, June 17, 1890.
" " " 492377, Febr. 21, 1893.
English " " 4757, 1891.
" " " 21696, 1892.
" " " 21707, 1892.

tube, into which he introduced hydrogen, coal-gas, or a chosen hydrocarbon. The apparatus served principally for making aluminum bronze from copper and corundum.

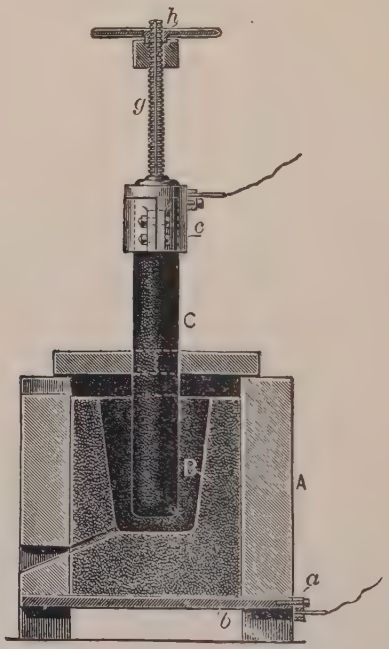

FIG. 23.

It is worthy of remark that the idea which is the basis of the Willson process—an idea dating from the year 1890—had been already developed by Minet in a patent of 1887, to which we shall return for a more detailed consideration later.

At about the same time Willson also constructed a calcium-carbide furnace, which, however, like

the Borchers furnace, was available merely for the production of carbide with an excess of carbon.

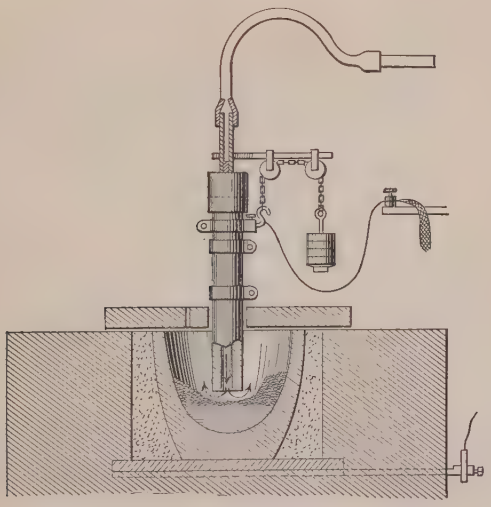

FIG. 24.

Moissan's Researches. — The attempts to reduce alumina in the electric furnace by means of carbon, which had been relinquished by Cowles, were resumed in the year 1892 by Moissan. He succeeded in producing at a high temperature an aluminum carbide, expressed in the formula C_3Al_4. Besides this, we owe to the labors of Moissan a great number of well-defined metal carbides, in particular the calcium carbide, which he produced in collaboration with Bullier. Moissan availed himself in its manufacture of a furnace which Fig. 25 shows in cross-section, and Fig. 26 in complete view.

The characteristic feature of this furnace, which was constructed in the year 1892 and exerimentally

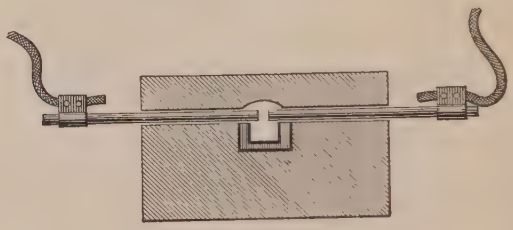

FIG. 25.

tested in the "Conservatoire des arts et métiers," are, according to Moissan's own statement, the

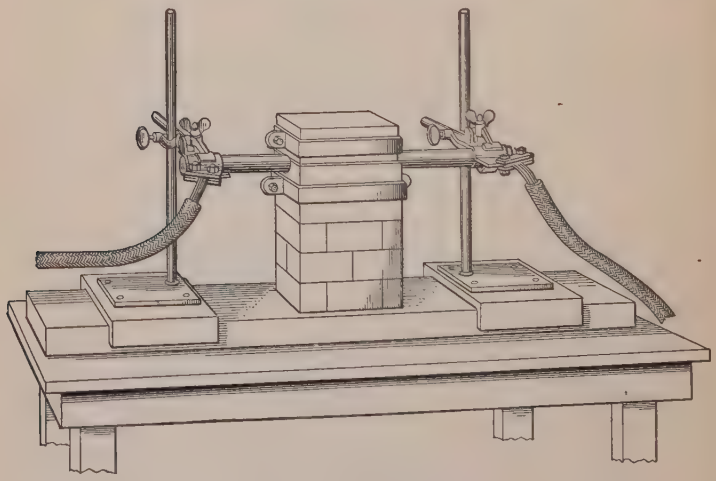

FIG. 26.

perfect separation of the electric and the thermic effects of the current, and the localizing of the developed heat to a space closed in on all sides.

The author, in the year 1891, built a furnace of a very similar type (Fig. 27), and, indeed, in the

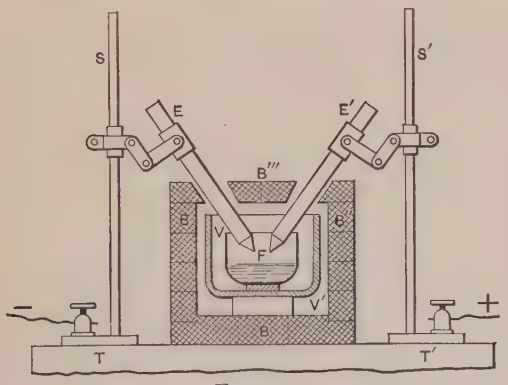

FIG. 27.

same laboratory with Moissan. Only the arrangement of the electrodes is different; in the case of the Moissan furnace they lie horizontally, while with Minet they may be fastened in any desired position by means of a special device.

Menges Process.* — This process makes use of the electric arc itself for the production of aluminum. Fig. 28 gives the entire view of the apparatus, as in use in 1886.

It resembles quite closely an arc lamp whose lower electrode is firmly fastened in the bottom of a crucible of good conducting material. The upper electrode pierces the lid of the crucible, and is kept by means of a mechanical arrangement at a convenient distance from the lower electrode. It

* D. R. P. No. 40354, 1886.

consists of a mixture of carbon, as a good conducting material, and the oxide to be reduced. The entire apparatus may be surrounded by a thick enveloping mantle, so that the melt may be undertaken under pressure as well. The process, however, has had no technical use.

Kleiner-Fiertz Process.* (1886).—This resembles, in general design, the Menges patent, except that it has special reference to the melting of cryolite. The electrodes reach into a vessel filled with cryolite, which has an inner lining of bauxite and clay (Figs. 29 and 30). Both are movable, but while the lower is to be adjusted by hand only, the movement of the upper electrode is governed automatically by means of a lever and a solenoid. The vibrations of the lever are deadened and limited by a piston, which, applied to the apparatus from above, dips into a speedily filled cylinder.

In consequence of its expensive mode of operation, the Kleiner-Fiertz furnace has been but little used.

Brin Brothers' Process (1888).—With this process it is possible, by means of a special device, to introduce into the arc light an indifferent gas, and so to have the reaction take place in an inactive atmosphere. The furnace pressure varies between 50 and 100 volts on the one hand, and between 20 and 25 volts on the other hand, and is governed according to the method by which the aluminum

* D. R. P. No. 42022, 1886.

is reduced. The brothers Brin propose two different methods. In either event, the original composition is the same; it consists of 100 parts of bauxite, 125 parts of common salt, and a certain quantity of borax.

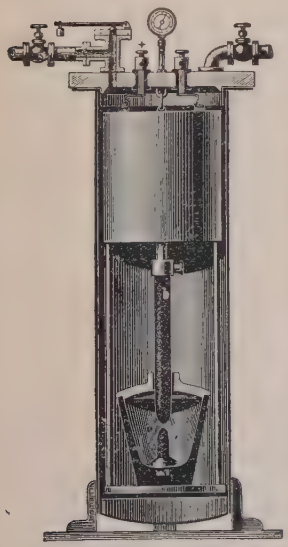

FIG. 28.

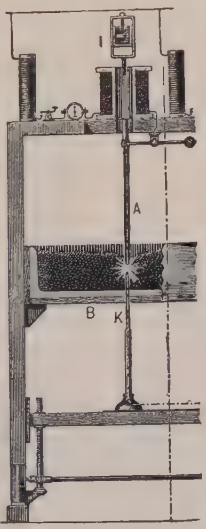

FIG. 29.

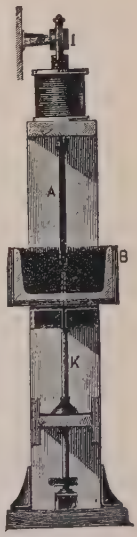

FIG. 30.

The first method operates exclusively electro-thermically. The specified materials are melted in a closed crucible until white fumes appear; the negative carbon electrode is then immersed in the mixture, while the positive remains at the surface of the bath. Under these conditions the arc works at a pressure of 50–100 volts. The aluminum is separated at the negative electrode; according

to the statement of the inventor, it would be mainly dispersed and lost, were the metal not protected from oxidation by a current of carbonic acid, which immediately conducts the fumes into the condensing-chamber.

In the second and electrolytic method, both carbon electrodes are dipped into the bath. At the positive pole chlorine is formed, from chloride of sodium; at the negative electrode sodium and aluminum are separated, the latter indirectly in consequence of the reduction of the alumina by sodium. Both metals are assembled on the bottom of the crucible in the form of an alloy rich in aluminum.

Bessemer Process. (Fig. 31).—The Bessemer fur-

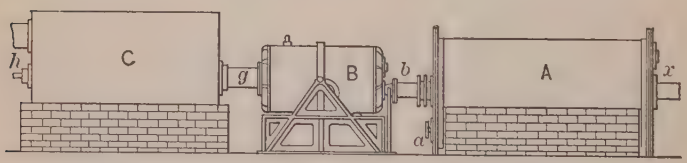

FIG. 31.

nace consists of three parts: the heating-chamber *A*, the reduction-chamber *B*, and the condenser *C*.

The heating-chamber *A*, of sheet iron, is filled with fire-brick, like the Siemens generators, and is calculated for a pressure of four atmospheres; at *a* the flame-gases, mixed with air, are introduced, while the products of combustion escape through the chimney at *x*.

When the bricks are brought to a red glow, the openings *a* and *x* are closed, and heating-gases are introduced through the tube *b* into the previously warmed reduction-chamber *B*; these gases, under the effect of a hot-air current which comes from *A*, and in consequence of the high pressure that prevails in *B*, are consumed at a very high temperature. When by this means the temperature desired in *B*—a red glow—has been obtained, the charge is introduced, which consists of a composition of powdered aluminum ore and carbon pressed into briquette form, to which as a flux soda, chalk, or borax is added. The whole is brought in the electric arc to a very high temperature, and thus the reduction of the alumina by carbon is effected. The aluminum-vapors arising are, with a simultaneous slackening of the gaseous tension, conducted to atmospheric presure in the condenser *C*, which is cooled with water.

Farmer Process.—Farmer produces pure aluminum directly in the electric arc, creating the arc within an electric crucible between two rods 10–15 mm in diameter. The rods consist half of carbon, half of corundum or emery, and are luted by means of sugar or petroleum residua. Into the crucible, through the tube *E*, air, coal-gas, petroleum-vapor, water-vapor, or zinc-vapor is introduced, whereby, according to the statement of the inventor, the reduction is accelerated; futhermore, it is said, the temperature of the crucible is heightened there-

by, and the dissociation introduced at the right moment.

K is a solenoid, which is applied to the principal circuit by means of a shunt, and is for the purpose of holding the arc firmly in the middle of the crucible. As soon as its resistance increases, the power of attraction of the magnetic coil K overcomes the elastic force of the spring k, the armature I sinks in consequence at i and, by means of a special contrivance seen in the drawing, causes the electrode carbons to approach each other.

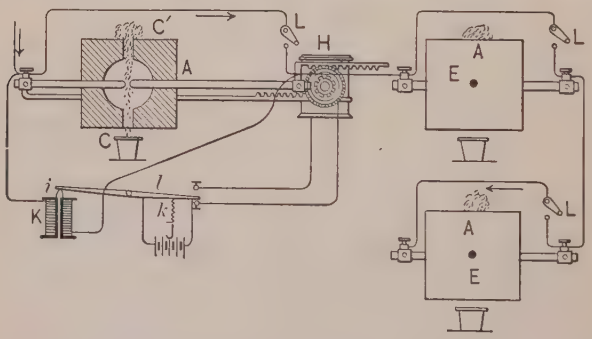

FIG. 32.

If the circuit-breaker L is set up in front of the crucible A, there is thus afforded a possibility of cutting out the corresponding crucible at the conclusion of the reaction, and so of keeping up the operation continuously.

The gases of reduction escape through the outlet C', while through the furnace-channel C what is, according to the statement of the patentee, almost chemically pure aluminum flows off.

The process is as adaptable to the direct as to the alternating current.

Gerard-Lescuyer Process.*—Fig. 33 gives the details of the furnace, which recalls one of the Johnson constructions. An arc is produced between

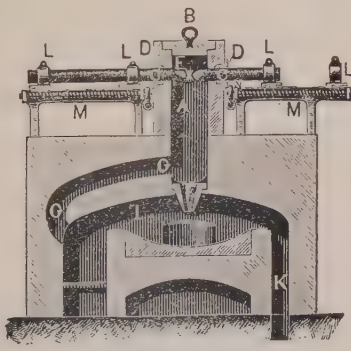

FIG. 33.

two easily exchangeable electrodes, which consist of a composition of 50 parts of dried alumina, 80 parts of carbon powder, and 100 parts of copper-dust, and are pressed into rods by the addition of tar or pitch. An endless screw makes possible the moving forward (in plain sight) of the electrodes, according to the amount of wear and tear.

The aluminum bronze formed falls on the bottom of a flame-furnace, and here comes into contact with lime, which accelerates the melt. The bottom is partially heated through the combustion

* D. R. P. No. 48040, 1887.

of carbon dioxide, which is conducted to the arc at *G*.

The metallic mass thus obtained, which contains about 20% aluminum, forms the point of departure for the production of pure aluminum; it is diminished and serves in the place of copper as an element of new electrodes. After repeated operations of this sort, by a continued process of enrichment, one finally succeeds in obtaining a metal almost pure.

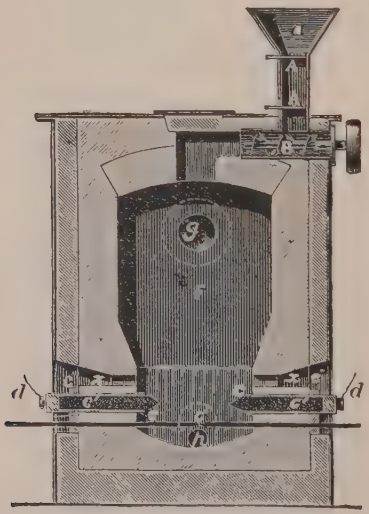

FIG. 34.

The Furnace of the Electric Construction Corporation* (Fig. 34) makes possible the heating of the charge by an arc or by a resistance interposed

* D. R. P. No. 55700, 1890.

between the electrodes. The charge itself may also, of course, serve as such resistance.

In Fig. 34 *F* is the furnace-pit with the charging-funnel *a*, which latter is provided with two slides *AA*, intended to prevent the entrance of air during the charging. On both sides of the smelting-furnace are the electrodes *c'c'*; these usually consist of carbon cylinders, which are enclosed in metal shells *cc*. *c''c''* are thin rods of carbon or metal which serve for heating when the circuit is closed. The gases and vapors developed pass off through the upper part of the furnace at *g*, while the slag is drawn off at *h*: *xx* are doors closed by means of clay plugs or clay mortar.

This furnace of the Electric Construction Cor-

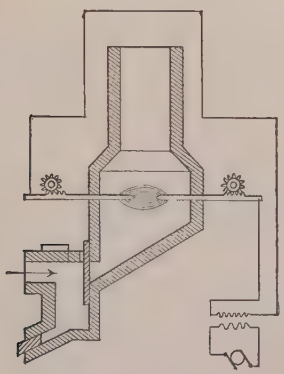

FIG. 35.

poration has been widely used in the production of aluminum by the electrothermic method.

Schneller and Astfalck Process (1890). — The temperature necessary for the reduction of the alumina is here developed by means of an alternating current transformed at a high tension (Fig. 35). The high tension is necessary because of the poor conductivity of the materials to be reduced. Simultaneously, and for the like reason, the wide upper surface which is exposed to the reducing-gases is used to advantage. For reducing-gases hydrogen or a convenient hydrocarbon is employed. The furnace-charge itself consists of alumina, aluminum sulphide, chloride, or fluoride.

(*b*) Electrolytic Processes for the Production of Aluminum.

In order to be able to subject a compound to electrolysis, it must first be converted into the fluid state of aggregation; this may occur through dissolving or melting. If the substance concerned contains water in chemical association, it might also be made fluid by melting in its water of crystallization or hydrate-water; however, for aluminum we have no examples of this kind. As for the separation of aluminum from the aqueous solutions of its salts, there are extant, indeed, several projects having this end in view. Yet, since they are all impracticable, we shall limit ourselves in the following pages to a brief description for the sake of completeness. The only electrolytic proc-

esses which have found a practical technical application depend upon the electrolysis of aluminum compounds in the molten condition.

The Electrolysis of Dissolved Aluminum Salts.—This subject has been thoroughly treated by Borchers in his *Electrometallurgy;** we give here a brief extract.

It is a fact experimentally verified that the electrolysis of aluminum salts in aqueous solution or in some other medium of solution containing hydrogen and oxygen, in consequence of the great affinity of the aluminum for oxygen in the nascent state, always gives the hydroxide, and never the metal; nevertheless some inventors have asserted that under certain conditions they have obtained metallic aluminum through the electrolysis of its dissolved compounds.

The oldest assertions of this sort are found in the English patent of Thomes and Tilly,† who electrolyze an aqueous solution of aluminum hydroxide freshly precipitated in cyanide of potassium, and in the patent of Corbelli,‡ who recommends the following electrolytes: 2 parts aluminum sulphate or alum, dissolved with 1 part calcium or sodium chloride in 7 parts water. The anode is to be quicksilver, the cathode zinc.

* Borchers, Elektrometallurgie, Verlag von H. Bruhn, Braunschweig, 1896, p. 108 ff.

† Engl. Pat. No. 2756, 1855. According to J. W. Richards, Aluminum, 2d Edition, London, 1890.

‡ Eng. Pat. No. 507, 1858. J. W. Richards, loc. cit.

Dingler's Journal, in Part I for August 1854, contains an account of an "alleged" process for plating copper galvanically with aluminum or silicon. In order to obtain aluminum, a solution of hydrate of alumina in hydrochloric acid is made; into the solution is introduced a porous vessel of clay, which contains an amalgamation of zinc plate and sulphuric acid in the proportion of 1 to 12. The zinc plate is connected by a copper wire with a copper plate of the same dimensions which is likewise immersed in the solution of alumina. After a few hours the copper plate should be covered with a thin incrustation of aluminum, which, it is stated, has the color of lead, becomes, on polishing, bright like platinum, and is tarnished neither in air nor in water. This metallic covering, it is alleged, is also precipitated from solutions of alum and aluminum acetate.

In a very similar apparatus silicon also is separated from an electrolyte which is produced by melting together 1 part silica with 2.5 parts sodium carbonate and dissolving the melt in water. If in addition a pair of Smee elements are connected with the circuit, the separation of the silicon very soon follows, and in fact, as the patentee, George Gore* of Birmingham, maintains, in the form of a silver-white incrustation.

J. Nickles† is of the opinion that in the pro-

* Philosophical Magazine, March 1854, p. 227.
† Journal de Pharmacie, June 1854, p. 476.

cesses just described an electrolytic result might clearly have been observed; that the precipitation, however, was by no means of aluminum, but rather zinc, which could have originated from the zinc sulphate contained in the porous cell.

Jeanson,* at a temperature of 60° C., electrolyzes aluminum salt solutions having a specific weight 1.15–1.16.

Haurd † recommends an aqueous solution of cryolite (?) in magnesium chlorides or manganese chlorides.

Bertram ‡ asserts that he has precipitated the metal from solutions of aluminum and ammonium fluoride.

J. Braun § (Berlin) affirms that he has produced aluminum at an ordinary temperature, by electrolysis of an alum solution, having a specific weight of 1.03–1.07.

According to an English patent of Overbeck and Niewerth, ‖ an aqueous solution of the salts of aluminum with organic acids is electrolyzed; or else, of compounds which form similar salts; or, finally, of aluminum sulphate in combination with other metal chlorides.

Senet ¶ claims credit for a process similar to

* Annual Record of Science and Industry, 1875. From J. W. Richards, Aluminum, 2d Ed., London, 1890.

† U. S. A. P. No. 228900, June 15, 1880. Richards, loc. cit.

‡ Compt. rend., Bd. LXXXIII, 1876, p. 854.

§ D. R. P. No. 28760.

‖ Engl. Pat. No. 5756, 1883; J. W. Richards, loc. cit.

¶ Cosmos les mondes, 1885; Richards, loc. cit.

the preceding; he employs only currents of 6–7 volts and 4 amp.

Walter * electrolyzes a solution of aluminum nitrate, using a platinized copper plate as electrode.

Reinbold † recommends the following electrolytes for the production of aluminum incrustations upon other metals: 50 parts alum are dissolved in 300 parts water, mixed with 10 parts aluminum chloride and heated to 93° C.; after the cooling down 39 parts cyanide of potassium are added to the solution.

R. de Montgelas‡ first precipitates from a solution of aluminum chloride containing iron electrolytic iron, and then, after the addition of lead oxide, zinc oxide, or tin oxide, separates the aluminum simultaneously with the metal of the added oxide.

By the process of Falk and Schaag § salts of aluminum are mixed with non-volatile organic acids in aqueous solution with the cyanides of copper, gold, silver, tin, or zinc; the conductivity of the bath thus obtained is increased by the addition of an alkali nitrate or alkali phosphate, and the resulting alloy then separated under the influence of the current.

Burghardt and Twining electrolyze aqueous solutions of alkali aluminates, to which cyanides and

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* D. R. P. No. 40626.
† Jewellers' Journal, 1887.
‡ Engl. Pat. No. 10607, 1886.
§ R. D. P. No. 48078.

eventually also other compounds of metal oxides and alkalis are added. At a temperature of 80° C., according to the statement of the patentee, aluminum or one of its alloys is precipitated.

According to Nansen and Pfleger,* in contrast to the previous processes, the cooling down of the electrolyte as far as to 40° C. directly promotes the separation of pure aluminum or its alloy with magnesium.

According to Rietz and Herold,† aluminum is precipitated from a solution containing aluminum, starch, and dextrose, which is electrolyzed between platinum electrodes of great current-densities; the aluminum being, of course, of a spongy consistency.

Scientific publications of American ‡ and German § origin contain accounts of a process of covering iron electrolytically with aluminum, which is said to have been practically carried out in the factory of the Tacony Iron and Metal Company, at Tacony, Pa.; more exact details, which from a metallurgical point of view would be interesting, are, however, lacking.

Felt proposes to separate aluminum in an apparatus which is depicted in Fig. 36, the purpose of which is very uncertain. *E* represents the cross-shaped copper cathode, over which a copper-wire

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* D. R. P. No. 46753.
† D. R. P. No. 58136.
‡ Iron Age, 1892, Feb. 25 and June 2.
§ Stahl und Eisen, 1892, Nos. 7 and 14.

netting G hangs from above. The positive electrode R, which is located in the upper part of the vessel, has the form of a circular grating, and consists of zinc, which is provided with quicksilver channels C for its amalgamation. A parchment diaphragm H divides the entire cell into two parts; the tube K serves as a means of emission for the

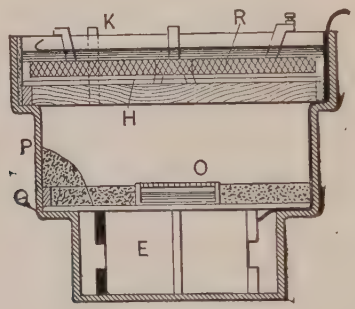

FIG. 36.

gases developed during the electrolysis. As an electrolyte dilute sulphuric acid is employed, which is mixed with some quicksilver nitrate dissolved in a hundred times its weight of water; from which the quicksilver found in the set-in vessel O is, last of all, constantly delivered. The aluminum ore P— of pure clay, for example—is fed upon the metal network, and, according to Felt, is decomposed into silica, which falls through the netting and is collected at the bottom of the apparatus, and into aluminum, which is precipitated upon the metal grating, not, however, upon the other parts of the cathode E. In order to obtain pure aluminum,

one of the surfaces of the netting is lacquered sufficiently so that the metal is only separated at the other surface.—It is needless to say that of this patent as well as of all others of this group no significant use has been made.

Finally, the process of Whole should be mentioned, by which a solution of alum, electrolyzed with the addition of cyanide of potassium, delivers metallic aluminum.

The Electrolysis of Molten Aluminum Compounds.—Among the countless processes which have for their object the production of aluminum by means of the electrolysis of its molten compounds, there are in particular but three which have found and still find technical employment, namely, those of Héroult, Minet, and Hall.

On looking through the descriptions of patents relative to our subject, these processes, both with regard to the fundamental idea as well as with respect to the composition of the electric bath and the arrangement of the apparatus, do not appear to differ materially from other processes of the same group. This was no doubt the reason why individual authors had doubts as to the practicability of the processes mentioned, and attained improvements that were perhaps too superficial, so that these processes offer nothing new. If one inspects them yet more closely, it is plain that the above-mentioned processes for the electrometallurgy of aluminum are quite novel,

and are related only very distantly to their predecessors.

If the electrometallurgy of aluminum was to be enabled to develop on a secure foundation, it was necessary above all to construct substantial apparatus, which must also show themselves capable of resistance to corrosive materials in a molten condition—for example, fluoride—and which could be harmed neither by the effect of the electrolyte nor by the length of an operation. Furthermore, it was necessary to secure carbon anodes which, together with the utmost cheapness and a slight percentage of impurities, united resistance to heat with a good conductivity; in short, the problem of a rational electrical furnace had to be solved.

Electrometallurgists were, then, in the presence of a very definite problem; it is true that as early as the beginning of the year 1887 electrolytic aluminum had been put upon the market, yet the processes actually capable of competition in the production of the metal date only from a later time—from the years 1890 and 1891; and through these methods, worked out with precision to the utmost detail, not merely was the aluminum industry promoted, but, together with this industry, electrometallurgy as a whole.

The furnaces which originally sufficed merely for the requirements of the electrolysis of melted compounds, as, for example, the furnace of Héroult, could later be used without material alteration

in electrothermic processes also, as in the production of alloys, of metal carbides, metal borides, and metal silicides.

Davy Process.—After Davy * had succeeded in dissolving the alkali hydrates by means of the electric current, he attempted in the year 1807 to apply the same method with alumina for the production of metallic aluminum; he reached, however, no result, owing to the weakness of the currents at that time available for the prosecution of his experiments. Not until a few years later,† when he resumed his investigations, did he succeed in producing an alloy of aluminum and iron, and then he did so in the following manner:

A platinum plate, which was connected with the positive pole of a galvanic pile of 1000 elements, was covered with a deposit of damp, hard-pressed alumina, into which an iron wire was introduced, which was in connection with the negative pole of the pile. The wire came forthwith to a red glow and melted at the point of contact. The mass was, when cooled down, more brittle than iron, and proved to be an alloy of iron and aluminum. Certainly the latter, in this reaction, was not separated by a purely electrolytic process, but rather by an electrothermic, which to some extent resembles the methods of Cowles and Héroult.

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* Philosophical Transactions, London, 1808.

† Ibid., 1810.

Bunsen Process.—To Bunsen * belongs the credit of being the first to produce pure aluminum by the electrolytic method (1854); and this he did by the electrolysis of melted aluminum compounds. He used for this purpose the apparatus which two years before had served him for the electrolytic production of magnesium (Fig. 37), and which

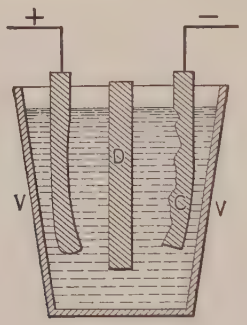

FIG. 37.

now rendered him notable service in dissolving the aluminum-sodium double chloride.

Deville Process.—Almost simultaneously with Bunsen, Henri St. Claire Deville † succeeded in obtaining small quantities of aluminum electrically; likewise through dissolving the aluminum-sodium double chloride.

Undoubtedly the French investigator, when on the 14th of August, 1854, he described his investigations before the Academy of Sciences, had as yet no knowledge of the researches of Bunsen, which

* Poggendorffs Annalen, XCII, 1854.

† Ann. de chimie et de physique, XLIII (1854), p. 27.

only shortly before, on July 9th, had appeared in Poggendorffs Annalen. But, even though he made no mention of this last publication of Bunsen, he did not omit in his communication to refer to the process by which Bunsen in 1852 had produced magnesium by the electrolysis of its chloride, and to acknowledge "that the German investigator has here indicated a method that might lead to interesting results in widely different directions."

The apparatus of which the French savant availed himself consisted of a porcelain crucible *P* (Fig. 38),

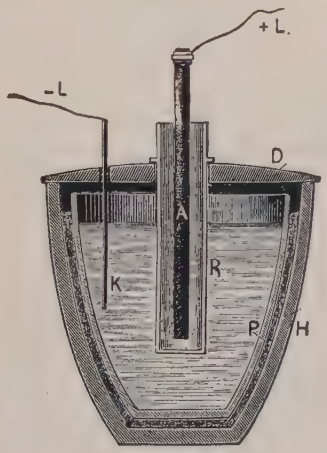

FIG. 38.

which was set into a Hessian crucible *H*; the whole was closed with a lid *D*, in which there were two openings, a small, slit-like one for the platinum sheet *K* serving as the negative electrode and a larger, circular one for the introduction of the

porous cylinder *R*. Into the latter was plunged the bar-shaped anode *A*, which consisted of retort-carbon. Between the bottom of this cylinder and that of the crucible was left a margin of several centimetres.

The crucible and the cell were filled to the same height with molten aluminum-sodium chloride, which was produced by heating a mixture of 2 parts anhydrous aluminum chloride and 1 part sodium chloride. These two salts unite at about 200° C. with a release of heat, whereby an easily flowing substance is produced which at the least rise in temperature gives off abundantly vapors of aluminum chloride.

With the passage of the current, the aluminum chloride is dissolved; the chlorine migrates to the anode *A* and there escapes. The aluminum separates off at the cathode *K*, whence from time to time it is removed, while the electrode is taken out of the bath and allowed to cool. In this way small quantities of aluminum are eventually obtained, in the form of a metallic residuum.

In order to keep the proportion of aluminum in the melt constant, Deville * recommends anodes which consist of a pressed mixture of aluminum oxide and carbon. This process was later taken up again by Le Chatelier and Lontin; I myself always added to my anode material some parts of

* H. St. Claire Deville, Aluminium, p. 95, Paris, 1859.

aluminum oxide, but less to prevent variations in the composition of the bath than to give an increased firmness to the anodes.

Gaudin Process.*—This depends upon the electrolysis of a fused mass of cryolite and chloride of sodium. By means of the current the fluoride is dissolved, fluorine is separated at the positive, aluminum at the negative pole. More exact details are not known.

Kagenbusch Process † (1872). — Clay is melted with the aid of fluxes, and, after the addition of zinc, electrolyzed; the zinc alloys itself, it is stated, with aluminum, and may be separated from the latter by distillation or by a winnowing process.

The patents of Berthaut ‡ (1879; similar to the process of Deville) and of Faure (1880), who dissolves aluminum chloride electrolytically, we here mention merely for the sake of completeness, without going further into a description of the details of the apparatus.

Lontin Process. — Even though the researches of this investigator of the question of aluminum production, which began before 1880 and ended only with his death (1886), reached no final conclusion, they are nevertheless of an abiding importance in electrometallurgy, in consequence of their

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* Moniteur scientifique, XI, p. 62, according to J. W. Richards, Aluminium, 2d Ed., London, 1890.

† Engl. Pat. No. 4811, 1872; Richards, loc. cit.

‡ Engl. Pat. No. 4087, 1879.

practicability and their precision—qualities which, indeed, characterize all of Lontin's operations; and if to-day we must acknowledge that the electro-metallurgy of our metal, thanks to the patents of Héroult, Hall, and Minet—which, moreover, are so similar that they seem almost to form one process,—rests upon a firm foundation, to Lontin belongs the credit of having been their predecessor.

In his researches (1882) he proceeded from the electrolysis of a melt in which alumina was dissolved, and which doubtless formed a mixture of cryolite and common salt, and hence had the same composition as, later, the baths of Héroult and Hall.

In 1886 Lontin presents his first proposal; it is true he retains the cryolite-salt melt; he does not, however, add to this alumina, but, reverting to the earlier arrangement of Deville, employs simply an anode which consists of a carbon-alumina compound. In the process Lontin supposes that the current in the electrolysis decomposes only the sodium chloride, and not the cryolite as well. The chlorine which separates at the positive electrode is united with Al_2O_3 and forms Al_2Cl_6 which mixes with the bath. As soon as the aluminum chloride has passed beyond a certain point in concentration, only the chloride and no longer the salt is decomposed by the current. From this moment aluminum is given off at the cathode, in a molten state, since the temperature of the bath is

higher than that of the metal, whlie at the anode the chlorine developed comes into reaction with the mass of alumina equivalent to the separated aluminum chloride.

It is true that Lontin, in these researches terminated all too soon by his premature death, reached no final result; yet, as we have already remarked, by his efforts the way to the goal was made plain.

Graetzel Process.*—This depends upon the electrolysis of a quantity of melted chloride and fluoride. The apparatus (Fig. 39) consists of a melt-

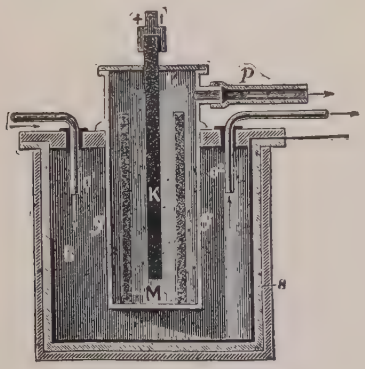

FIG. 39.

ing-vessel of porcelain, stoneware, or some other suitable fire-resisting material, which is protected from direct contact with the fire-gases by a metal mantle. The vessel has an inner lining of metal, preferably aluminum, which serves as a cathode.

* D. R. P. No. 26962.

The anode is formed by a carbon rod *K*, which is enclosed in a porcelain tube *G*, provided with slot-openings *g* and a tube *p* for the release of the chlorine.

During the electrolysis a reducing-gas is conducted through the apparatus, entering at *O'* and escaping at O^2. In order to lessen the pressure, and in order simultaneously to add to the bath fresh material according to the rate of exhaustion, there are in the porcelain tube *G*, at both sides of the carbon electrode and not having any contact with it, plates or bars *M*, which consist of a composition of equivalent quantities of alumina and carbon.

This process, which may be classed with the Lontin process for aluminum, has never been technically applied, and even its inventor, according to the statement of Borchers, in his capacity as director of the Hemlinger Aluminum and Magnesium Works has made use, in obtaining aluminum, not of his own patent, but of the patent of Beketoff, which depends upon the reduction of cryolite by means of magnesium.

Boguski-Zdziarski Process* (1884).—This patent had for its object principally the production of aluminum alloys. Its similarity to the Lontin method may be seen from the subjoined description of the patent: Cryolite or other aluminum

* Engl. Pat. No. 3090, 1884.

compounds were mixed with the appropriate fluxes and smelted in an iron or graphite crucible heated by flame-gases. On the bottom of the crucible the metal is found which is alloyed with aluminum. The cathode during the electrolysis is the alloy itself, while a bar of carbon dipped into the melt serves as anode.

Farmer Process * (1885). — This rests upon the electrolysis of molten aluminum chloride in a crucible whose conducting walls form the cathode.

Grousilliers Process † (1885). — In order to avoid the very considerable loss of aluminum chloride by evaporation, owing to the high temperature of the electrolytic cell, Grousilliers recommends electrolysis under pressure in closed vessels.

Grabau Process. ‡ —Among the impurities of aluminum produced electrolytically from molten fluorides, we have to take into account principally those that—like iron and silicon—in consequence of the more or less strong effect of the bath upon the walls of the vessel owing to the temperature-relations and the contained fluoride, are among the last to be successfully melted.

With cooled pole-cells Grabau, therefore, hopes to obtain pure aluminum by the following process: In the electrolytic dissociation of a molten bath of cryolite and common salt, we know that chlorine

* U. S. A. P. No. 315266.
† D. R. P. No. 34407.
‡ D. R. P. No. 45012.

is separated at the positive, molten aluminum at the negative pole. Since molten cryolite affects every fire-proof, non-conducting material, the parts of the apparatus concerned must be protected by an invulnerable insulating covering from the effect of the bath or of the elements separated therefrom. Grabau attains this end by means of the following device (Fig. 40).

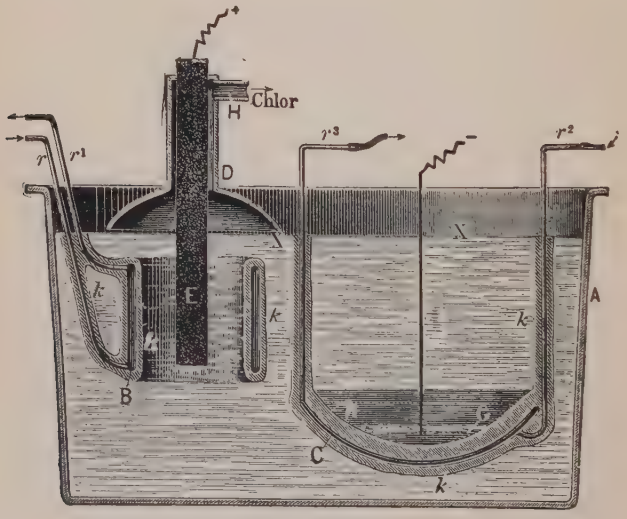

FIG. 40.

A is an iron melting-vessel, which is heated from the outside by fire-gases to such a degree that the molten material remains in an easily flowing state; its level mounts to *XX*. *B* is a double-walled metal cell of ring-shaped cylindrical form, which is cooled with air or water. The flowing aluminum

separated is assembled in a trough-shaped collecting-vessel C, which is likewise provided with double walls, between which air or water circulates. In consequence of the cooling thus brought about the molten mass congeals on the entire surface of the cell, of the collecting-vessel and the conducting-tubes rr' and r^2r^3, and the non-conducting crust K formed hereby cannot be attacked either by the smelting or by the aluminum.

In my opinion, an apparatus of this sort is not practicable; at least, it would not accomplish the object intended by the inventor, since he has undoubtedly overlooked the fact that the iron outer vessel A is affected by the melt. Iron salts will be formed which mingle with the bath, and according to the proportions of their composition are dissolved by the current, so that the aluminum precipitated under these conditions will always contain a large percentage of iron. In order to obtain better results, the vessel A would have to be cooled to the same degree as the cell B and the containing vessel C; in that event, however, the heat from outside would fall off, and the additional warmth necessary for the production of the molten flow would be taken away from the work of the current.

Héroult's First Process* (1886).—By this patent, similar to the first Lontin process, a solution of alumina is subjected to electrolysis in molten

* Engl. Pat. No. 7426, 1887 —Henderson-Mandataire.

cryolite, whereby a quantity of alumina equivalent in amount to the metal separated is continually replaced. The crucible *A* (Fig. 41), which is heated

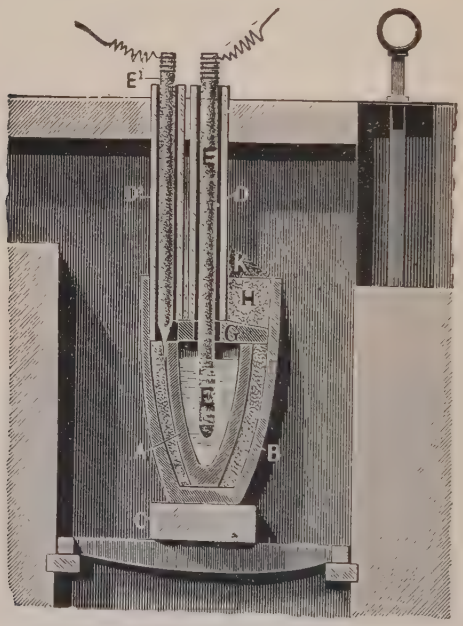

FIG. 41.

from without, consists of carbon and, at the same time, forms the cathode. It is surrounded by a second crucible *B* of graphite, which serves as a protecting envelope; the space between these two crucibles is filled with graphite powder. The contact with the external circuit is brought about by the carbon bar *E'*, which is enwrapped by the clay tube *D'*. The carbon anode *E*, which is like-

wise protected by a clay tube *D*, is introduced into the bath through an opening in the roof *G*. The latter is covered with a deposit of alumina. The entire apparatus rests upon a support of fire-clay. For the electrolysis an electromotive force of 3 volts is sufficient.

The patent of Henderson, according to his description, is similar to the majority of processes that have actually passed into technical employment; it seems to me, however, that its introduction in practice has not been seriously contemplated even by the inventor.

Lossier Process.* — A composition of melted cryolite and sodium chloride is electrolyzed under a gradual addition of silicate of alumina or kaolin. In this way, however, pure aluminum cannot be obtained, but merely a metal containing a large percentage of silicon.

According to a process similar to one of the Lossier patents—the addition of bauxite to a bath of aluminum fluoride—the author has succeeded under very advantageous conditions in obtaining a ferro-silicon aluminum which may be employed directly for the refining of steel; this process, however, seems as yet to have met with no favor among metallurgists.

In conclusion we may mention the Rogers †

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* D. R. P. No. 31089.

† Proceedings of the Wisconsin Natural History Society, April 1899. Richards, loc. cit.

process, which depends upon the electrolysis of molten cryolite with cathodes of melted lead; then the patent of A. Winkler * (Görlitz), the idea of which is, however, conceived from a point of view wholly erroneous, since the patent recommends the electrolysis of alumina phosphates and borates; and finally the

Feldmann Process † (1887), according to which a composition of aluminum-sodium double fluoride with barium chloride, strontium chloride, calcium chloride, magnesium chloride, and zinc chloride, or else (1889) a mixture of aluminum haloid salts with the oxides of electropositive metals, is electrolyzed.

Among all the processes thus far explained for the electrolytic separation of aluminum, there is found none which is of industrial insignificance. Of technically important processes there are but three, namely, those of Minet, Héroult, and Hall. These will be described in detail in the following pages.

Minet Process.

I had a twofold aim in my researches upon the electrometallurgy of aluminum.

On the one hand, I sought to find, for a metal with such a promising future, the most economical method of production; on the other hand, I wished

* D. R. P. No. 45824.
† D. R. P. No. 49915, 1887.

to solve a problem of much more universal significance—the problem of the electrolysis of anhydrous, molten electrolytes.

We know that the electric current is able to operate in two ways—electrolytically and electrothermically. The researches of the author have been chiefly of an electrolytic nature, though in many of the furnace constructions electrothermic principles have been involved.

Since in my investigations I made the universal characteristics of the electric current my point of departure, I came straightway upon its capacity—of such great technical significance—for storing up large amounts of energy within a limited space; a principle which may perhaps be expressed as follows: No matter by what method, from a physical point of view, an electrical phenomenon may be produced, whether it is an effect of light or of heat, an electrolytic or an electrothermic process, the utilization of the energy assembled by means of the electricity is the greater, the smaller the space in which the reaction takes place.

I ascribe it solely to the fact of my adhering as closely as possible to this principle, whose correctness, indeed, is evident from what has been already said, that I was able through my efforts, which date from February 1887, to solve the problem of the electrolytic production of aluminum from its molten salts. It is, however, the same principle upon the basis of which Cowles and

Héroult succeeded in producing electrothermically the copper and iron alloys of aluminum, and which Moissan and Bullier were later to apply in order to obtain crystallized calcium carbide; and if Willson was able to produce only slightly defined carbides, this was largely because of a neglect on the part of this investigator to heed the principle I have mentioned.

The electrolytic decomposition of fire-melted materials had found but a limited application until there was a successful electrometallurgical process of producing aluminum. This method of decomposition was employed under certain circumstances in chemistry for scientific purposes—for example, in order to effect the decomposition of material reducible with difficulty; and though the same method was also proposed for the production of certain metals, such as the alkalies and alkaline earths, the investigations in this domain were limited, at least so far as regards any industrial application.

In my efforts to bring upon the market electrolytic aluminum as cheaply as possible, and in large quantities, I have at the same time sought to find the best conditions for the electrolysis of molten salts in particular. With reference to this question we must consider: the composition of the bath, its temperature, movement, density, unchangeableness, constancy, the dimensions of the electrodes and of the crucible which contains the melt, and

finally the nature of the various parts of the apparatus. That these results could be reached only through special devices and by the use of new apparatus goes without saying.

In the proof of a theoretical interpretation of the observed phenomenon one obtains a formula which unites the constants of the current with those of the electrolyte, for the three stages of the beginning, the course, and the conclusion of the electrolysis.

The first part of my researches occupied two years; during this time my method was used industrially in two places, in Paris, Impasse du Moulin-Joli (1887), and in Creil (1888); later the establishment in Creil, where I could avail myself of only one 30-horse-power steam-engine, was transferred to Saint-Michel de Maurienne (1891), where I worked up to the year 1894 with a water-power of 500 H.P.—For the means of carrying out my experiments I have to thank the Bernard brothers.

Choice of the Electrolyte.—There are three kinds of aluminum salts, which in a molten condition may be subjected to electrolysis: the haloid salts, that is to say, those in which the acid radical is a halogen; the oxy- or double salts, consisting of aluminum oxide, combined or mixed with an aluminum-halogen salt; and finally, according to the statement of some inventors, the sulphides. I myself, in continuing the investigations of Deville and

Lontin, concerned myself chiefly with the chloride and the fluoride of aluminum.

The melting-point of the pure fluoride lies at a very high temperature, and indeed so near the boiling-point that upon heating it passes directly from the solid into the gaseous state. But even if one could keep it molten for a sufficiently long time, it would nevertheless not allow itself to be electrolyzed, since, like all pure salts, it is a poor electric conductor in the molten state; in order to increase its conductivity it must be combined with the salt of another metal, for example, sodium fluoride—that is to say, a double salt must be formed.

Aluminum chloride melts at a low temperature (185° C.), and shows with regard to melting the same appearance as the fluoride; like the fluoride, also, it is not a good conductor until it becomes a double salt.

For electrolyte I used the following compounds: on the one hand 40 parts aluminum-sodium double chloride and 60 parts sodium chloride, on the other hand 40 parts aluminum-sodium double fluoride and likewise 60 parts sodium chloride.

The aluminum chloride is also uncommonly volatile as a double salt, even when it is mixed with an excess of alkali-salt; at the slightest rise in temperature corrosive vapors arise from the bath, which make more difficult the control of the electrolysis and are not without attendant risk.

A bath with aluminum fluoride as the chief element gives the best result.

Properties of the Electrolyte.—The composition of the melt, as I finally decided upon it, is expressed in the formula

$$12NaCl + Al_2F_6.6NaF.$$

Its melting-point lies at 675° C.; at 1056° C. it begins to evaporate; its density at 829° C. amounts to 1.76; its coefficient of expansion in the molten state is 5.10^{-5}; its specific conductivity at 870° C. is 3.1, and therefore its specific resistance is 0.323 ohm. If C_t is the specific conductivity and R_t the specific resistance at the temperature t, we have the formula

$$C_t = 3.1[1 + 0.00334(t - 870°)],$$

$$R^t = \frac{0.323}{1 + 0.00334(t - 870° \text{ C})}.$$

For a current strength of 4000 amp. the charge amounts to 60 kg. At 800° C. the melt is sufficiently fluid to maintain the electrolysis uninterruptedly, and at the same time is so slightly volatile that the losses in consequence of evaporation during 24 hours do not exceed 5%.

The bath unites in itself the best possible conditions for work. If its properties—its melting-point, its density at 870° C., etc.—are compared with the corresponding properties of metallic aluminum, the truth of the assertion will appear that

we meet with scarcely another instance in metallurgy where there is such a favorable coincidence.

Since aluminum, as we know, melts at 625° C., it is already separated in a fluid state by electrolysis, at the bath-temperature of 870° C.; since, furthermore, its density (2.63) is considerably higher than that of the electrolyte (1.76), the metal simply flows off along the cathode and is assembled at the bottom of the crucible, whence it may be drawn off through a tapping-vent.

How widely the electrometallurgical production of aluminum differs in this respect from that of the other metals, the following comparison will show. As the average density of the substances subjected to the electrolysis in a molten state—potassium, sodium, magnesium, lithium, beryllium—we may take 1.75. Since, now, the density of potassium is 0.87, of sodium 0.97, of lithium 0.59, the metals mentioned, in contrast to aluminum, are not assembled at the bottom of the crucible, but rise upward, where by means of special contrivances they must be united and prevented from oxidation by air. With magnesium and beryllium the metal-density and the bath-density are not materially different (1.76 and 1.73 as compared with 1.75); the metals, therefore, remain floating in the melt, and are assembled only in a form of the apparatus constructed for the purpose.

As for the temperature of the melt, the relations in the majority of electrolytic baths are in so far

favorable, that the temperature generally, without impairing the ready movement of the electrolyte, is sufficiently far below the boiling-point of the metal concerned, but above the melting-point.

Decomposition-voltage of the Electrolyte. — Upon the passage of the current, the aluminum chloride is first dissolved, since this chloride, of all salts present, has the lowest decomposition-voltage.

Name.	Equivalent Formula.	Heat of Formation, Cal.	Decomposition-voltage, Volts.
Aluminum fluoride.....	$Al_{2}/_{6}F$	70	3.04
Sodium chloride.....	NaCl	97.3	4.23
Sodium fluoride.....	NaF	110.8	4.82

The heat of formation of aluminum fluoride, it is true, has not been experimentally determined, yet it may be estimated, when we take into consideration the following facts: Upon comparing the other halogen salts of aluminum (chloride, bromide, iodide) with the corresponding potassium haloids, one finds between the pairs of homologous members an equal and constant difference of 51.75 cal. One may therefore justly assert that between the heats of formation of aluminum fluoride and potassium fluoride there exists a like difference. Now, the heat of formation of potassium fluoride has been experimentally determined to be 118.1 cal., and therefore the heat of formation of aluminum fluoride amounts to $118.1 - 51.75 = 66.35$ cal. A wholly analogous comparison between the aluminum

and the hydrogen haloids gives for the heat of formation of the aluminum fluoride 73.7 cal. If we take the average of the two figures, we obtain for the desired heat of formation 70 cal.

The decomposition-voltage is reckoned by the universal formula

$$e = 0.04346\ C,$$

if C is taken to signify the equivalent heat of formation of the electrolyte. For aluminum fluoride we obtain, then (see Appendix, page 218).

$$e = 0.04346 \times 70 = 3.04 \text{ volts},$$

which value is introduced into the table above.

Electrolysis. — The electric current brings about the decomposition of the aluminum double salt into aluminum, which is separated at the negative pole, and into chlorine, which is formed at the positive electrode, while the released sodium fluoride remains unaltered in the melt. The process is expressed in the following equation:

$$Al_{2/6}F.NaF = Al_{2/6} + F + NaF.$$

Upon the passing through of 96435 coulombs, the reaction takes place according to the stoichiometric quantities expressed in this equation.

Should it finally be desired, according to the scale on which the decomposition takes place, to enlarge the bath by the addition of fresh quantities of cryolite, the melt would be so greatly enriched

with sodium fluoride that the latter substance would at once be present in excess, so that one would then obtain in the electrolysis, not aluminum, but sodium. This, indeed, is capable of experimental verification. In order to avoid this result the two following methods may be adopted.

Regeneration of the Bath by means of Aluminum Fluoride.—Into the bath while the electrolysis is going on aluminum fluoride is introduced in quantities equivalent to the sodium fluoride released, so that the percentage of aluminum in the electrolyte remains constant. The aluminum fluoride added unites with sodium fluoride according to the equation

$$Al_{2/6}F + NaF = Al_{2/6}F.NaF.$$

To every regenerated g-molecule there corresponds, at the negative pole, 2/6 g-atoms of aluminum, while at the positive pole a g-atom of fluorine escapes.

Regeneration by means of Alumina.— The regeneration follows as a consequence of the fact that during the electrolysis alumina is deposited in the form of a fine powder in the neighborhood of the anodes. There are two principal hypotheses as to the reaction that takes place in this case.

1. The alumina, whether it unites with the released sodium fluoride or is simply dissolved in the molten mass, is electrolyzed simultaneously with the aluminum fluoride, since its equivalent heat of

formation (65 cal.) very nearly approaches that of the aluminum fluoride. In what form the alumina is contained in the melt is, as we have said, not demonstrated. It may on the one hand unite with a molecule of aluminum fluoride for an oxide-fluoride of the formula $Al_{2/6}O_{1/2}.Al_{2/6}F$, whereby a molecule NaF is released; on the other hand it may also form directly an oxide-fluoride of the composition $Al_{2/6}O_{1/12}.NaF$.

Héroult and Hall, who accept these hypotheses, are of the opinion that it is almost exclusively alumina which is decomposed by the electric current. At the negative electrode aluminum is freed; at the positive pole oxygen separates off, which violently attacks the electrode-carbon. The process is expressed in the equation

$$Al_{2/6}O_{1/3} = Al_{2/6} + O_{1/3}.$$

It is a fact that the anode in time becomes much corroded; and this effect is in proportion to the quantity of aluminum separated off.

2. According to a second hypothesis, it is accepted as a fact that the work of the current limits itself exclusively to the aluminum fluoride. According to this conception, the alumina serving for the regeneration is converted at the anode, by the fluorine there developed, into aluminum fluoride, according to the formula

$$Al_{2/6}O_{1/2} + F = Al_{2/6}F + O_{1/2};$$

the aluminum fluoride formed unites with the free sodium fluoride to form a double salt:

$$Al_{2/6}F + NaF = Al_{2/6}F.NaF,$$

and oxygen escapes. (See Appendix, page 218).

I consider this second hypothesis the more probable. In its favor, moreover, is also the circumstance that the fluorine, in its effort to develop at the anode, is not completely absorbed by the added alumina, so that, if the composition of the bath is to be kept constant, in addition to alumina interchanging quantities of aluminum fluoride must be added to the melt.

Through the successive addition of a composition of common salt and aluminum-sodium double fluoride, in the proportions given above, the losses due to evaporation are compensated for, and the level of the bath is kept at the same point.

Electrolytic Constants. — Of the three constants here coming under observation, the counter-electromotive force e, the resistance ρ, and the potential difference at the electrodes E, we have already defined e; it remains therefore to examine more closely the two other quantities, ρ and E.

The resistance ρ varies with the proportion of double salt contained in the bath, and with the dimensions of the electrodes.

The four baths A–D in the following table had the same composition—70 parts common salt and 30 parts double fluoride, with an insignificant percent-

TABLE I.

Bath.	Temperature, Deg. C.	Resistance, Ω.	Dissolving-tension, Volts.
A	900	0.0044	2.4
	1000	0.0033	2.3
	1100	0.0025	2.17
B	870	0.024	2.50
C	870	0.0012	2.50
D	870	0.0071	2.50

age of silicon and iron, which, however, at the beginning of the operation was speedily removed by the electrolysis itself; merely the size of the electrodes was different in the four baths.

The potential difference E was determined at three different times: before, during, and after the electrolysis.

1. *Measurements before the Beginning of the Electrolysis.*—The bath *B*, in which the experiments were begun, was covered with a melt of the above mentioned composition; the percentage of silicates and iron salts amounted to about 2% of the entire mass.

In the melt electrodes of different materials were dipped and their surfaces measured; anode and cathode were always of the same size. After the reading of the temperature the electrode-potential was obtained either by the condenser method, in which the condenser was discharged into a highly sensitive galvanometer of the Lord Kelvin con-

struction, or by the compensation method with a Lippmann electrometer for the zero instrument.

The electromotive force thus determined proved to be in the majority of cases very small; frequently in the course of a measurement it changed its sign; the maximum positive and negative values are given in Table II. It may be seen from this table

TABLE II.

POTENTIAL DIFFERENCE BETWEEN THE ELECTRODES BEFORE THE BEGINNING OF THE ELECTROLYSIS.

Electrodes.		Submerged Surface, Sq. Cm.	Temperature, Deg. C.	Potential Difference E between the Electrodes, Volts.
Anode.	Cathode.			
Copper.....	Copper.....	104	824	$\left\{\begin{matrix}+0.0025\\-0.0019\end{matrix}\right.$
Copper.....	Fresh carbon	104	824	$\left\{\begin{matrix}+0.002\\-0.002\end{matrix}\right.$
Platinum...	Platinum...	8	920	$\left\{\begin{matrix}+0.0056\\-0.0012\end{matrix}\right.$
Polarized carbon *.....	Fresh carbon	200	824	0.22
do.	Iron.....	200	824	0.32
Fresh carbon.	do.	200	824	0.25
Polarized carbon.....	Molten aluminum.....	200	824	$\left\{\begin{matrix}1.95\\1.70\\1.50\end{matrix}\right.$

* By polarized carbon is understood one that has already served as anode in the normal electrolysis of aluminum fluoride, and is therefore loaded with oxygen.

that the electrodes, in so far as they consist of copper, platinum, or fresh carbon, do not occupy the place of some important or constant electromotive force; E even changes its sign; its maximum value in both cases is about the same: it amounts between copper-copper and between

copper–fresh carbon to about 0.002 volt; with platinum it varies between +0.0056 and −0.0012 volt.

If the positive electrode consists of polarized carbon, the negative of fresh carbon, the electromotive force, which at the beginning of the measurement in the open circuit is not inconsiderable, decreases rapidly, as soon as the electrodes are united by an external resistance, and is finally zero. If, on the other hand, the negative electrode consists of iron or molten aluminum, the positive of fresh or polarized carbon, the system directly forms a galvanic element which remains electromotively effective for some time after the closing of the circuit.

2. *Measurements during the Electrolysis.*—These were carried out during four separate periods (*a*, *b*, *c*, *d*), which were distinctly characterized by different current-densities (current-strengths per sq. dm); and they will be described in the following pages. Both electrodes, in these tests, consisted of pressed carbon.

Period a.—The melt *B*, whose resistance ρ at 870° C. amounted to 0.024 Ω, served as electrolyte. The entire surface of each electrode was 4.25 sq. dm. Even with very weak electromotive forces, which were delivered from an external current-source, the current which passed the electrolyte was easily measurable; after breaking the circuit the two electrodes showed only a slight polarization,

which differed but little from that measured before the passage of the current.

From the current-density 0 to 0.01 amp. per sq. dm, E, the difference of potential between the electrodes. is visibly proportional to the current-intensity, Table III gives the values obtained in comparison with those calculated. In our example E is reckoned for the given current-densities according to the equation

$$E = kI = 3.1I.$$

TABLE III.

DIFFERENCE OF POTENTIAL DURING THE PERIOD a.

Current-strength, I	Current-density, $d = \frac{I}{4.25}$	Difference of Potential between the Electrodes, Volts.	
		Obtained.	Calculated.
0.0021	0.0005	0.0067	0.00651
0.0055	0.0013	0.017	0.0171
0.0189	0.0042	0.055	0.0558
0.0260	0.0060	0.077	0.0804
0.0425	0.0160	0.128	0.133

In explanation of the above formula, with its coefficient k, unusually large in comparison with the resistance, we may remark that the latter is composed of two terms: of ρ, the resistance of the electrolyte, and of the term k', which refers to the electromotive force of the polarization. We have

$$k = \rho + k',$$

and therefore

$$E = \rho I + k'I.$$

Period b.—For current-densities from 0.02 to 2 amp., the difference of potential E is no longer proportional to the current-strength, but increases more slowly than the current-density. I was unable to obtain a general comparison between E and I for this period.

Period c. From a current-density of 2 amp. per sq. dm to 100 amp., the values E and I stand in the following relation, given according to the principal formula:

$$E = e + \rho I.$$

This equation was thoroughly proved by experiments; I have found it verified in a great number of baths, in which I varied the dimensions of the electrodes and the temperature, while the composition of the bath was constantly maintained.

I give two examples below. The first has reference to the bath *B*, which showed a small percentage of silicates in its contents. The temperature during the test was kept constantly at 870° C. The second example refers to the bath *A*, which contained neither impurities nor silicates; the temperature in this case was varied between 900° C. and 1100° C.

In the first case, the original proportion of silicates, in consequence of the electrolysis, gradually diminished, and soon fell to zero. I could clearly perceive five separate phases; the resistance remained constant during them all, being equal,

namely, to 0.024—a proof that the percentage of silicate is without effect upon this value. The current-strength was kept during the described experiment between 10 and 400 amp., corresponding to a current-density of 2.36 and 94 amp. per sq. dm.

From the following table may be seen the increase of the decomposition-voltage with the continuance of the electrolysis, for 870°.

TABLE IV.

Phase.	$E=e+\rho I$.	Phase.	$E=e+\rho I$.
1	$1.33+0.024I$	4	$1.95+0.024I$
2	$1.50+0.024I$	5	$2.50+0.024I$
3	$1.75+0.024I$		

During the first period e was 1.33 volts. As long as the melt still contained significant quantities of silicates, this value of e remained constant, an indication that during this time the work of the current is limited to the dissolution of the silicates. In the mass, as the proportion of silicate decreased, e increased, and finally reached 2.50 volts; at this stage scarcely any traces of impurities were still found in the electrolyte, so that 2.50 volts may be accepted for the decomposition-voltage of the aluminum fluoride.

In practice, one only goes with the anode current-density as far as to 50 amp. per sq. dm, and the electrodes are taken of such dimensions that the resistance of the bath is lessened in proportion as

the current-strength utilized increases, so that the current density retains its value permanently. Thus the resistance of the bath C, which was calculated for a current of 4000 amp., amounted merely to 0.0012 ohm, in order to obtain a current density of 50 amp. Since the counter-electromotive force amounted to about 2.5 volts, the value of E was derived from the formula

$$E = e + \rho I = 2.5 + 0.0012.4000 = 7.3 \text{ volts.}$$

Likewise I found in the bath B, with the same current-density and the current-strength of 200 amp. dependent thereon, the following value for the difference of potential E:

$$E = e + \rho I = 2.5 + 0.024.200 = 7.3 \text{ volts.}$$

If the temperature was kept constant during the preceding experiment, special measurements at the bath A had reference directly to the influence of the temperature upon the tension of the cell. The melt A was subjected to three different temperature-points of the electrolysis, and each time ρ and e were determined. That the values calculated from these quantities according to our formula closely agree with those experimentally determined, the following table will show.

The entire surface of each group of electrodes that was sunk in the bath A remained constant during the three tests and was equal to 50 sq. dm.

TABLE V.

900° C. $E=2.4+0.0044J$.			1000° C. $E=2.34+0.0033J$.			1100° C. $E=2.17+0.0025J$.		
Current-strength, J, Amp.	Difference in Potential, E.		Current-strength, J, Amp.	Difference in Potential, E.		Current-strength, J, Amp.	Difference in Potential, E.	
	Measured, Volts.	Calculated, Volts.		Measured, Volts.	Calculated, Volts.		Measured, Volts.	Calculated, Volts.
196	3.26	3.26	572	4.23	4.23	152	2.48	2.55
403	4.12	4.17	650	4.48	4.48	598	3.65	3.67
485	5.05	4.97	910	5.30	5.54	1010	4.90	4.48
885	6.18	6.25	1013	5.78	5.74	1430	5.74	5.74

The current-density was varied between the limits $d=\frac{152}{50}=3.04$ and $d=\frac{1430}{50}=28.6$. Furthermore, for an electrode-surface of 100 dm^2 and a current-density up to 40 amp., I found, in confirmation of our fundamental formula, a very good agreement between the calculated and the measured values; and this, indeed, even at a current-strength of 4000 amp.

Period d.—If the current-density exceeds 100 amp. per sq. dm, the difference in potential at the electrodes may no longer be expressed as a simple function of the values e and J. It then quickly attains the electromotive force of the arc, about 30 volts; simultaneously there is a bright gleam of light around the anode, and this heat-effect of the current disappears only with the falling-off in the current-density.

3. *Measurements after Breaking the Circuit.*—If, after breaking the circuit, the electrodes are united by means of a metallic resistance, a polarization-current flows in the outer closed circuit, which perhaps in the course of an hour assumes a constant value and lasts until the melt has cooled down, that is to say, has hardened.

Table VI refers to the bath *D*, whose resistance was about 0.0075 ohm. After a current of 800 amp. had flowed through it for a while, the latter was suddenly interrupted, and the cell at the same time closed by a metallic conductor whose resistance *R* was chosen about ten times as large as that of the bath, so that

$$R = 10\rho = 0.075 \text{ ohm.}$$

The decrease in potential *E* and the current-strength *I* in the outer circuit are given in the following table:

TABLE VI.

DISCHARGE OF THE BATH *D* AFTER BREAKING THE CIRCUIT.

Exterior Resistance $R = 0.075$ ohm.

Minutes Afterward.	Difference in Potential E_1.	Current-strength, $I = \frac{E_1}{0.075}$
0	1.95	25.3
5	1.88	25
15	1.58	21
30	1.38	18.4
45	1.29	17.2
60	1.20	16
75	1.20	16

TABLE VII.

RESISTANCE ρ DURING THE DISCHARGE.

Test-periods.	External Resistance.	Potential Difference E_1 and e_1	Current-strength I.	Bath-resistance, reckoned according to the Formula $\rho = \frac{e_1 - E_1}{I}$.	ρ Average.	ρ General Average.
1	Open circuit	1.72	0		0.0076	0.0071
	0.075	1.58	21	0.0081		
	0.0125	1.10	88	0.00705		
	Open circuit	1.72	0			
2	Open circuit	1.26	0		0.0067	
	0.0125	0.795	64	0.0072		
	0.0083	0.715	86	0.0063		
	0.0050	0.550	110	0.0065		
	Open circuit	1.26	0			
3	Open circuit	1.17	0		0.0070	
	0.125	0.755	60.4	0.0068		
	0.0050	0.480	96	0.0072		
	Open circuit	1.17	0			
4	Open circuit	0.825	0		0.00735	
	0.0125	0.510	40.8	0.0070		
	0.0050	0.343	68.6	0.0077		
	Open circuit	0.825	0			

If the current of polarization is interrupted for an instant, in order to close it again immediately, and if at each interruption one measures the electromotive force e_1 between the electrodes with the circuit open, one finds in time a distinct lessening of e_1. The resistance ρ of the electrolyte may be determined by the formula

$$e_1 = E_1 + \rho I, \quad \text{or} \quad \rho = \frac{e_1 - E_1}{I},$$

and must be identical with that which is calculated by the principal formula, $E = e + \rho I$, or $\rho = \frac{E - e}{I}$.

Table VII refers to the melt D, whose resistance during the electrolysis was found to be 0.0075 ohms. After the breaking of the primary current circuit, the electrodes in turn were short-circuited by resistances of 0.075, 0.0125, 0.0083, and 0.0050 ohm, and the current-strengths in each one of these resistances measured at different times; furthermore, the fall of potential at the electrodes was determined both with the open (e_1) and the closed (E_1) circuit.

Table VII shows that the resistance of the melt in the periods of discharging and charging is remarkably similar (0.0071 as contrasted with 0.0075 ohm).

Verification of the Faraday-Becquerel Law. — The amount of aluminum, which theoretically is separated by a coulomb, amounts to 0.0936 mg; the ampere-hour (3600 coulombs) yields, therefore, 0.337 g of aluminum. In practice, however, inasmuch as the negative electrode consists of carbon, scarcely more than 80% of the theoretical value is obtained. This is caused by the fact that the aluminum, according to the proportion of its composition, is partly reunited with the halogens (chlorine, fluorine) separated at the positive electrode, and is thus retained in the electrolyte.

If, however, iron cathodes are employed instead of carbons, the theoretical yield is easily obtained;

P. S. THE DETROIT, MICH.

I obtained, for example, with a current of 4000 amp. during twenty-two hours, that is to say, then, by means of 88,000 ampere-hours, 30 kg aluminum, while the theoretical value was

$$P = 88000 \times 0.337 = 29656 \text{ kg.}$$

The aluminum unites in this case with the cathode-metal to form ferro-aluminum, an alloy which—with a proportion of 7% of iron—is affected by the bath far less than pure aluminum.

The difference of potential with carbon cathodes amounts to 7.5, with iron cathodes to 7 volts.

In the following statement, for both cases, the electrical energy is calculated which is consumed by a current of 4000 amp. during twenty-two hours, in order to determine from this and from the quantity of metal separated during this time the expenditure of energy per kg aluminum.

Pure Aluminum.

Production P during 22 hours: 23.75 kg.

Electrical energy in horse-power hours:

$$\frac{7.574 \cdot 000 \cdot 722}{9.81 \cdot 75} = 900 \text{ HP}^{h}.$$

Expenditure of electrical energy T per kg aluminum:

$$T = \frac{900}{23.75} = 38 \text{ HP}^{h}.$$

Ferro-Aluminum.

Production P in 22 hours: 29.656 kg.
Electrical energy in horse-power hours:

$$\frac{7.4000 \cdot 22}{9.81 \cdot 75} = 840 \text{ HP}^{\text{h}}.$$

Expenditure of electrical energy T per kg aluminum:

$$T = \frac{840}{29.656} = 28 \text{ HP}^{\text{h}}.$$

Requisite Nature of the Apparatus. — The crucible, which contains the electrolytes, must be so constituted that it will not be affected by the melt, since, by any such effect, on the one hand the purity of the aluminum would suffer, on the other hand the crucible would soon become unfit for use. From an economic standpoint also, if it is desired to obtain pure aluminum, it is absolutely necessary to limit to a minimum the twofold attack which the crucible suffers from heat and from the molten salts, or, if possible, to do away with it entirely.

It is known that impure aluminum is more easily corroded by chemical reagents than pure metal; in addition, there is the unfavorable circumstance that while pure aluminum shows normal wear with the effect of reagents, thus permitting the durability of objects made of the metal to

be approximately determined beforehand, impure aluminum is so irregularly affected that it has but a limited usefulness. The impurities of aluminum electrolytically produced consist principally of silicon, iron, and traces of carbon; while the amount of these was originally 1%, at present it hardly exceeds 0.15%. In this connection I subjoin some very instructive analyses:

	1890	1893	1898
Silicon	0.90%	0.25%	0.02%
Iron	0.40%	0.40%	0.12%
Aluminum	98.70%	99.35%	99.86%
	100.00%	100.00%	100.00%

It is seen that the percentage of silicon with increasing perfection in apparatus grows notably less, so that it finally becomes altogether negligible, while the percentage of iron has become less, but yet remains always considerable. This peculiar phenomenon depends upon the origin of the admixtures mentioned; for while the silicon comes principally from the melt, the various additions and the electrode-carbons—and hence from materials which it is possible to obtain in any degree of purity desired—the iron comes from the crucible and its armature, and may be avoided only by means of certain devices which we shall now proceed to describe more in detail.

Technical Furnaces.—Of the three furnaces of

Minet's construction, we shall describe, to begin with, the

First Type.—The apparatus (Fig. 42) consists of a cast-iron smelting-vessel *VV* of parallelopipedal form, which is externally protected by brickwork from the attack of the superheated gases; for the bath is in this case maintained in a molten state not merely by the current heat, but also by artificial heat from an external source of warmth. If the addition of warmth from without decreases, and if the calories necessary for the melt are furnished by the current alone, the brick covering serves principally for the purpose of protecting the furnace as much as possible from cooling off through radiation.

For electrodes, carbon bars are employed; the cathode *C* is constructed directly over a crucible *cc*, which consists of the same carbon material as the cathode, and is for the purpose of taking up the aluminum that flows slowly down at the cathode. In order to avoid any effect of the bath upon the melting-vessel, and thus to prevent the melt taking up iron salts, which would all be more easily decomposed by the current than aluminum fluoride, the crucible walls are connected to the cathode as a shunt-derived circuit with the aid of a resistance *R*, which is so dimensioned that through it flows only 5% of the whole current. By means of this device the inner walls of the melting-vessel are protected from every attack, since along these

walls an extremely thin layer of aluminum is precipitated, which is renewed incessantly. With this furnace, which was devised in the year 1887, I have obtained an aluminum which contains only

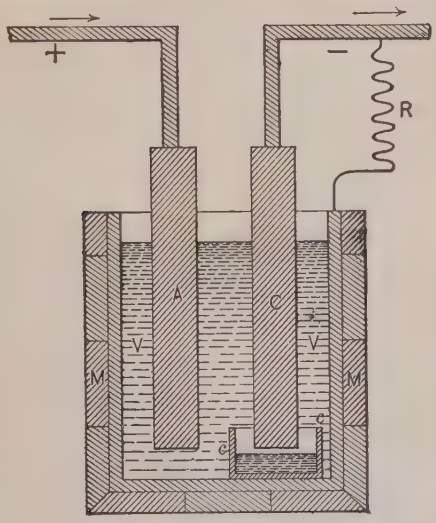

FIG. 42.

0.51% of impurities, namely, 0.33% of silicon and 0.18% of iron.

Nevertheless this apparatus can lay no claim to technical utility, since the metal vessel *V* is subject to very rapid waste, on the one hand from the aluminum which is precipitated upon its inner surface and which penetrates the walls, while it forms an alloy, ferro-aluminum, easily melted in comparison with cast iron; on the other hand, from the heating-gases which circulate about the crucible. The wasting is of such a sort that after

an eight or ten days' course the melt already trickles through, and the vessel must be permanently set aside.

Second Type.—In this furnace the melt is brought about solely by the current heat. The crucible consists of metal, and is likewise of parallelopipedal form. It is lined inwardly by a layer of carbon, which serves as negative electrode (Fig. 43). The anode consists of one or more blocks of carbon, which are arranged in the middle of the crucible.

At the beginning of the operation the percentage of iron separated is very small; it may sink to 0.1–0.2%, since the crucible material (usually cast

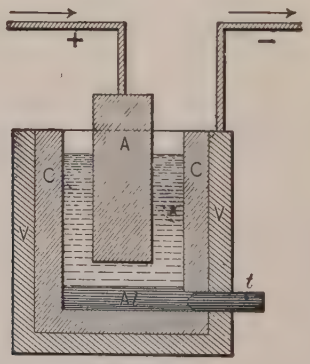

FIG. 43.

iron) in consequence of the lining we have mentioned does not at first come in contact with the melt. At the high temperature, however, to which the carbon mantle is brought (750–850° C.), this soon becomes porous, allows the melt to trickle

through, and as quickly brings about a contact between the bath and the metal crucible. From this moment the latter is in electrolytic connection with the anode *A*, and in consequence aluminum is separated thereat, and in addition sodium, which, on account of the considerable proportion of sodium chloride in the bath, is precipitated simultaneously with aluminum, provided the electromotive force at the electrodes exceeds the decomposition-voltage of common salt, 4.35 volts.

If the sodium is separated exclusively at the surface of the carbon covering *CC*, this brings with it no further disadvantage. The metallic sodium reduces the aluminum fluoride coming into contact with it, and thus frees an equivalent quantity of aluminum. If, however, the sodium is formed in the pores between the wall of the crucible and the carbon mantle, in its immediate neighborhood there is not found a sufficient quantity of aluminum fluoride to exchange with the sodium to form sodium fluoride and aluminum. Hence the sodium penetrates into the mass of carbon saturated with aluminum fluoride, corrodes it, and reduces it finally to powder.

The separated aluminum is assembled at the bottom of the crucible, and is drawn off through the tapping-vent *t*. At first almost free from all impurities, it becomes ferruginous with continued operation. Many apparatus of this type keep in

condition for thirty to forty days; others become almost immediately useless.

If the aluminum is destined for alloys, this furnace may be employed to advantage industrially, at least in this way: by making the crucible of that metal, or of one of those metals, entering into the alloy. The absorption of the crucible material into the melt during the electrolysis in this case, of course, is attended by no inconveniences.

Third Type.—This stands midway between the first type and the second. The addition of heat is provided for entirely by means of the current. The metal crucible (Fig. 44) is also in this case

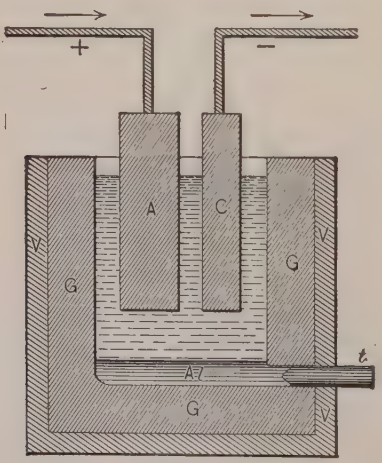

FIG. 44.

enveloped in a carbon covering, the strength of which, however, is far more significant than in the case of the preceding furnace; and the covering

is entirely independent of the electrodes. The aluminum flows away along the cathode *C* and is collected in a basin which is set in the middle of the bottom of the crucible. From this basin the metal may be drawn off through the channel *t*.

Since in this apparatus the covering does not take the place of an electrolytic process, it gains, to an unusual extent, in durability and constancy. The same thing is true also of the metal crucible wall, which may be cooled in such a way that the temperature of its inner surface remains lower than that of the melt, so that the latter cannot penetrate so far as to the walls of the crucible. Since, furthermore, there is no sort of electrolytic connection between crucible and anode, the melt remains free from a percentage of iron, and we at once have the condition for the production of very pure aluminum.

If aluminum is to be used for alloys, the crucible may, as in the case of the second type of furnace, be made suitably of one of the metals with which the aluminum is to be alloyed.

Furthermore, with a heavy lining and thorough cooling from without, the temperature may be lowered to below 500° C., while that of the melt amounts to about 750° C.; under these conditions it is very easily possible to make the crucible of aluminum, so that one is able to obtain a metal which for its sole impurity shows, at most, traces of silicon.

The space occupied by the furnace is not much greater in the case of the third type than in that of the second. If, in order to avoid certain phenomena of heat, the anodes are of such dimensions that only a current of 50 amp. per dm^2 of surface passes through, there is free play, at least in so far as the cathode is concerned—though the current-density must not be so great that there is an excessive heating of the cathode; the current-density at the negative electrode may be something like ten times as great as at the positive, amounting, therefore, to about 500 amp. per dm^2. We see that the construction of the cathode demands no considerable increase of volume for the third type in comparison with the second.

It should be remarked, also, that the cathode remains for a long time capable of being used; frequently I have had cathodes in use for eight days before they required to be renewed. The anodes, on the other hand, must usually be renewed twice a day; still, this is no more true of the furnace of the third type than in the case of that of the second type; so that the last-described construction unites in itself all the requirements of an industrial apparatus.

Technical Data.—If an external source of heat is employed, in order to keep the bath in a molten state—this is true only of the first and second types—the difference in potential at the electrodes amounts to 5–6 volts; if, however, the operation

is carried on without an addition of heat from the exterior, the electromotive force varies between 7 and 8 volts.

Table VIII gives the electrolytic data within further current-variations.

TABLE VIII.

Date of the Test.	Constitution of the Cathode.	Duration of the Test in Hours	Current-intensity in Amperes	Difference of Potential at the Electrodes in Volts.	Weight of the Metal Separated off in Grams.		Efficiency $\phi = \frac{p}{P}$
					Obtain'd p	Theoret. P	
The addition of heat is partially the consequence of heating from without. First type of furnace.							
1887							
May 7	carbon	15	89	5·5	250	455	55%
July 13	"	14	90	4	260	428	60%
Sept. 27	iron	23	100	5·5	400	782	51%
Nov. 26	carbon	12	142	5·75	380	579	75%
1888							
Feb. 4	carbon	13	180	6	500	796	62%
Aug. 4	"	12	360	6	1000	1460	68%
1889							
Sept. 30	carbon	20	700	5.6	2600	4760	54%
Nov. 20	"	20	800	5.6	2800	5440	52%
Dec. 5	iron	20	800	5·5	3600	5440	66%
1890							
Jan. 20	iron	7	975	6.1	1900	2320	82%
Dec. 10	carbon	22	1500	5·55	6500	11220	58%
The addition of heat is entirely the consequence of the work of the current. Second type of furnace.							
1892	carbon	24	3000	8.25	16157	24480	66%
	iron	24	3000	7·75	20074	24480	82%
1893	carbon	24	3500	8.25	19421	28560	68%
	iron	24	3500	7·75	24276	28560	85%
1894	carbon	24	4000	8.25	22848	32640	70%
	iron	24	4000	7·75	29376	32640	90%

In all these tests the difference of potential at the electrodes was kept approximately constant

by means of corresponding alterations in the furnace- and electrode-dimensions. With a current-interval of 89–1500 amp., in the case of the first type of furnace—during the simultaneous partial employment of an external source of heat—the electromotive force varied between 4.55 and 6.35 volts, the efficiency ϕ between 52 and 75% in proportion as carbon cathodes were used, and between 51 and 82% in proportion as iron electrodes were used. With the second type the electromotive force during a current-interval amounted to from 3000–4000 amp., and with the avoidance of any external heating to 8.55 volts with carbon cathodes and 7.75 volts with iron cathodes—hence with an unlined furnace-chamber—while the efficiency reached in the first case 70%, in the second 90%, of the theoretical value.

Expenditure of Energy per Kg Aluminum. — First type of furnace; a portion of the necessary heat being taken from an exterior heat-source.

Carbon Cathodes.

November 26, 1887. ..... 35 horse-power
December 10, 1890. 40

Iron Cathodes.

December 5, 1889. ..... 33 horse-power
January 20, 1890. ..... 30 "

Second type of furnace; the heat is exclusively caused by the current.

1894.

Carbon cathode..... 42.5 horse-power
Iron cathode..... 30.5 "

If we draw the inference from what has been said, we see that the expenditure of energy for the two furnaces is about the same; it increases slowly, however, with increasing strength of current.

Héroult Process.

The various furnace-constructions of this investigator we have already described (see pages 30–32). As for the electrolytes, Héroult employs a melt of aluminum-sodium double fluoride (cryolite) without any addition whatever of a salt of the alkalis or alkaline earths. The aluminum-salt decomposed by the electrolysis is replaced by anhydrous alumina which is added to the melt during the operation, mixed with some parts of cryolite.

At the Metallurgical Congress which was held at Paris during the World's Fair of 1900, in a debate on the subject of the production of aluminum, Héroult took the floor in order to make a communication regarding his first researches, after some consideration of the historical aspect of the subject. I will give the description of this portion of his achievements in his own words:

"We may say that the first thought of a technically practicable process for the electrometallurgical production of aluminum originates from the year 1886. Bunsen's and Deville's successful at-

tempts in the electrolytic decomposition of aluminum chloride had made a profound impression; thanks to the labors of Favre and Silbermann and to the researches of Berthelot, there were also at hand, even at that time, very reliable thermochemical data; and hence it needed but a step—for we have already reached a period subsequent to the invention of the dynamo-machine—in order to arrive at the point where we stand to-day; although, to be sure, the original materials for the manufacture of aluminum—alumina and cryolite—were already universally known.

"In view of these considerations, I became convinced that the electrolytic production of aluminum was only a question of time. I next attempted such a production with aqueous solutions; since, however, these experiments all resulted in failure, I passed immediately to the electrolysis of molten halogen-salts.

"We must not forget that the electrical industry was then only in the first stage of its development. To procure carbons having a diameter greater than 50 mm was at that time not yet possible. The few crucibles available in laboratories and factories were produced by hollowing out rotating retort-carbon.

"After countless failures, I once observed that in an attempt to electrolyze melted cryolite the iron cathode became fissured, allowing the contents of the crucible to flow out. Considering the temperature

with which I then worked, and the current, which I took from some Bunsen elements, I could not understand how it was that iron should melt under these circumstances. A careful investigation of the remains of the cathode led me to suppose that an alloy might have formed, and when, some days thereafter, I sought to lower the temperature of the electrolyte by the addition of aluminum-sodium double chloride, I was able, to my surprise, to state it as a fact that the carbon anode gave clear indications of having been attacked. I concluded from this that an oxide was here operating, the reduction of which must have taken place at the expense of the anode. I verified this conjecture, and found it was actually so—that the aluminum-sodium double chloride contained considerable quantities of alumina, which originated from the dissolution of the chloride brought about by moisture. The way was now indicated by which a technically practicable aluminum-process might be obtained. The matter was always more difficult than one would suppose; I will, however, pass lightly over the details of further attempts, which do not radically differ.

"My practical knowledge of chemistry was at the time that of a student of twenty-three; of special knowledge I had as good as none at all. Under these circumstances, it is needless to say that after I had taken out my first patent I sought the counsel and encouragement of those men who

were then considered authorities on this subject. Pechiney (Salindre), whom I first approached, explained to me that aluminum was a metal of restricted usefulness; at most, it might be used for opera-glasses; and whether I wanted to sell the kilogram for 10 or 100 francs, I would not be able to dispose of one kilogram more. It was otherwise in the case of aluminum bronze, of which considerable quantities were handled commercially, if I could produce it cheaply; I would then, beyond a doubt, come out even in my reckoning.

"I had already in this connection undertaken some successful experiments; and I therefore laid aside for the time being the production of pure aluminum and turned to a series of new researches, which in the year 1887 led to a second patent.

"In this additional patent a system of electric furnaces and a process were described which made possible a continuous production of alloys of aluminum, and particularly of all metals difficult to melt and reduce.

"Although in point of time the electric furnaces of Siemens and Cowles anticipated my invention, my furnace had yet other special features, such as the tapping-vent, etc., which passed into universal electrometallurgical use. I will only mention that, for example, all carbide factories have introduced carbon crucibles with a movable electrode and a tapping-vent.

"When my investigations had reached this

stage, I went to Switzerland, where I concerned myself for a year almost exclusively with aluminum bronze. I soon saw, however, that the difficulty lay not herein, but in the production of pure aluminum, and so, in conjunction with Dr. Kiliani, I took up again the process of 1886.

" I may here be permitted to state my attitude with regard to the vexed question of the theory of the reaction. Several investigators are of the opinion that the electrolysis does not consist in the dissolution of the alumina, in contradiction to my patent, in which I expressly speak of the electrolysis of the alumina. I have been able to demonstrate that the alumina may be electrolyzed, since I succeeded in fusing it in the arc and in decomposing it by the continuous passage of the current.

" It is true that I obtained only a slight yield of metal (some hundred grams); still, every error in this connection is excluded, and we must, therefore, of necessity infer that in my experiments simply the electrolysis of alumina has taken place. In fact, if we dissolve pure cryolite by means of the current we obtain pure aluminum, not, however, fluorine also. The latter, with sodium fluoride, which, indeed, is present in excess in consequence of the decomposition of the aluminum fluoride, forms a composition which is still constant at the temperature concerned. This may be demonstrated by grinding the cooled mass and digesting it with water. There is thus obtained an indissoluble

portion, which shows all the peculiarities and the composition of cryolite, and a soluble part, which is nothing but acid sodium fluoride. If, however, the operation is carried on at a higher temperature, one does not obtain aluminum, but sodium, which develops in abundant vapors.

"From these experiments we must reach the conclusion that sodium primarily is separated at the cathode through the electrolysis, which then in its turn reduces aluminum fluoride in a molten state. In this case, therefore, we have only aluminum at the cathode. If the temperature, on the other hand, is increased, sodium-vapors are developed at the negative electrode, and the reduction of aluminum fluoride does not take place. On the basis of this hypothesis, the rôle of the alumina may easily be explained. We have, on the one hand, a molten compound, which contains fluorine in excess; on the other hand, alumina and carbon. If now the heats of formation of alumina and aluminum fluoride are of a like magnitude, we have in favor of the transposition of the oxide the circumstance that oxygen is freed hereby, which, as soon as it comes into contact with the anode, attacks the latter."

Use of the Héroult Patents. — Héroult establishments are found in France, Switzerland, Germany, Austria, and England. In France is the "Société Electrométallurgique Française," which turns to account the Héroult process. This society was

founded by Gustave Munerel, and has notably developed under the direction of Emile Vielhomme. In Paris M. Dreyfus is its representative. The director of the establishment in La Praz is Victor Arnould. The society possesses two factories with water-power, one in Froges (Isère), the other in La Praz (Savoy). In both establishments carbon electrodes and electrolytic aluminum are produced. It also possesses a factory for chemical products in Gardannes (Bouches du Rhône), where alumina is produced.

For Switzerland, Germany, and Austria the Héroult patents are in the possession of the Aluminium-Industrie-Aktien-Gesellschaft at Neuhausen, which in Neuhausen (Switzerland), in Rheinfelden (Baden), and in Lend-Gastein (Austria) operates according to this process. The company obtains its alumina from the Bergius factory in Silesia.

In England it is the British Aluminum Company which has obtained the license for the Héroult patents. To it belongs an establishment (water-power) in Foyers (Scotland), and an alumina factory in Larne-Harbour (Ireland); also factories in Greenock and Milton-on-Trent, where electrodes and aluminum plates are made.

Among the persons who in a technical or a financial way have promoted the work of Héroult may be mentioned: Gustave Naville, superintendent of Escher Wyss, and Colonel Huber of the Oerlikon machine factory, who together established the

Schweizerische Metallurgische Gesellschaft, and have erected the first aluminum factory in Switzerland (Neuhausen, near Schaffhausen); also Dr. Kiliani, Frei of Neuhausen, and factory-superintendent Schindler.

The English company was founded by Ristori, with support on the part of Sir William Thomson (later Lord Kelvin).

C. M. Hall Process.

This process resembles both of the preceding ones. In contradistinction to the Héroult patent, the aluminum-sodium double fluoride is not added in its pure state, but, as in the Minet process, mixed with changing quantities of salts of the alkalis and alkaline earths. As admixtures we have to take into account the chlorides of potassium, sodium, lithium, and the fluorides of sodium, lithium, or calcium. These various admixtures are for the purpose of keeping down the melting-temperature of the electrolyte, in order to maintain an easily flowing bath at a lower temperature.*

Hall has constructed a great number of apparatus, which may be divided into three general groups. Fig. 45 presents the vertical section, Fig. 46 the complete view of a furnace of the first type, in which, according to the statement of the inventor, aluminum is to be produced by the electrolysis of

* American Patent of July 9, 1886. American Patents 400766 and 400664 of April 2, 1889.

a solution of alumina in sodium- (or potassium-) aluminum double fluoride.

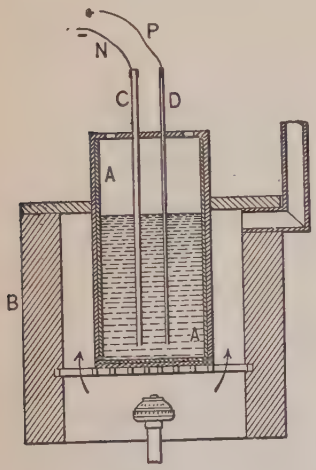

FIG. 45.

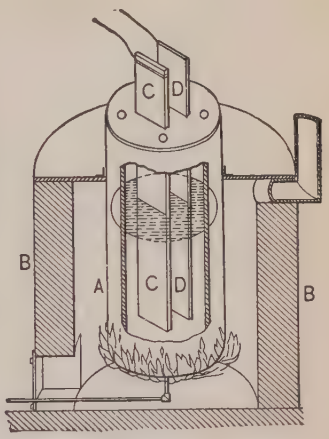

FIG. 46.

The electrodes *C* and *D* are of carbon; the vessel *AA*, which contains the electrolyte, consists of clay or steel; it is lined within by a layer of carbon, which protects it against the corroding attack of the molten alumina.

Figures 47 and 48 reproduce two other furnace types of the Hall construction; in Fig. 47 both electrodes are separated by a partition; in Fig. 48 this is not the case.

In a third group of Hall furnaces the melting-vessel forms at the same time one of the electrodes (Figs. 49 and 50).

All these apparatus were originally arranged for

external heating; so far as I am aware, however, the arrangement has of late years been abandoned.

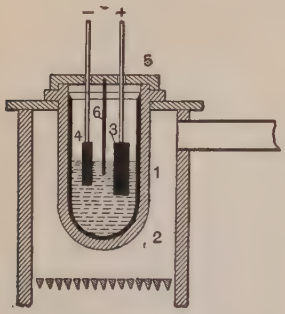

FIG. 47.

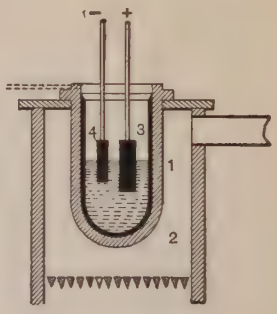

FIG. 48.

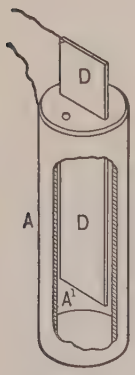

FIG. 49.

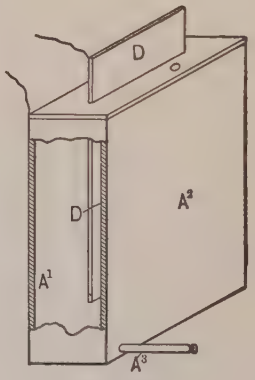

FIG. 50.

The Hall process is employed in Pittsburg, Penn., and in Saint-Michel-de-Maurienne, France.

In the following pages several additional processes will be cited, which, it is true, have not been employed industrially, but which—particularly as

regards the arrangement of separate details of the apparatus—are not without interest.

J. B. Hall Process.—The iron crucible is here lined on the inside with carbon, and serves as the cathode. For the electrolyte a melt of the chlorides of aluminum, sodium, and lithium is employed. The aluminiferous anode placed in the middle of the apparatus provides for the renewal of the alumina.

Berg Process.—A mixture of aluminum ore (for example, cryolite and bauxite), carbon and alkali nitrate (or alkali sulphide) is electrolyzed at a low temperature. The nitrate or sulphide is to bring about the separation of the aluminum created from the alumina by reduction with carbon, from the accompanying impurities, iron, silicon, etc., which latter arise from the matrix of the ore or from the crucible material. The less oxidizable aluminum is not affected thereby.

Bull Process (Fig. 51).—*A* and *B* are two graphite crucibles, connected one behind the other, which are heated by means of a gas-firing. In the crucible *A* common salt or chloride of potassium is melted, in *B* aluminum chloride is volatilized. The melt is subjected to electrolysis in crucible *A*, wherein the positive electrode is formed by the crucible walls, the negative by the graphite rods *EE*.

The course of the operation is as follows: Under the effect of the current, sodium is formed in crucible *A*, upon which the aluminum-chloride vapors arising from *B* are allowed to react. The reaction

is very lively; the aluminum is assembled at the bottom of the crucible over a layer of pure alumina a', and is in this way protected from the effect of the chlorine vapors developing only at the upper portion of the crucible walls. The metal is drawn off every four or five days; the chlorine is con-

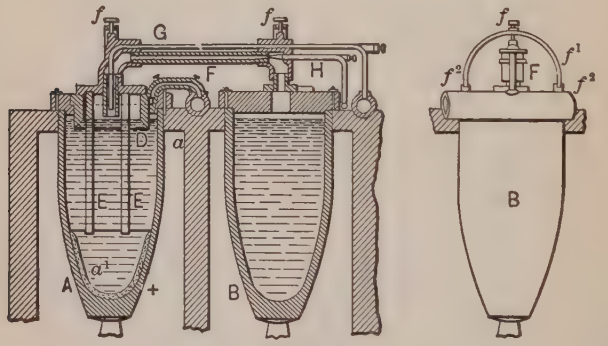

FIG. 51.

ducted by means of a peculiar tube, seen in the illustration, into a collecting-tube in which a jet of steam maintains the circulation.

In order to force the aluminum chloride vapors from B to A and to accelerate their reduction, a jet of hydrogen is introduced at H, which is generated by passing water-vapor over sodium and thus decomposing it. From this point the operation is so carried on that in the crucible A more sodium is formed than is necessary for the decomposition of the chloride of aluminum coming from B. The sodium-vapors are conducted through the tube G to T, where they are condensed.

Daniel Process. — This process differs from Deville's only in the respect that here, in consequence of the regeneration of the aluminum-sodium double salt, a continuous operation is provided for. Fig. 52 gives the details of the crucible which contains the molten aluminum chloride, Fig. 53 the entire view. The crucibles *B*, which are charged with the aluminum

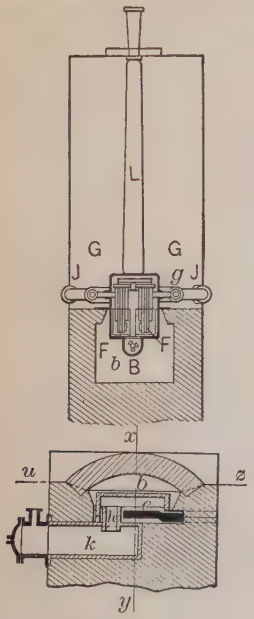

FIG. 52.

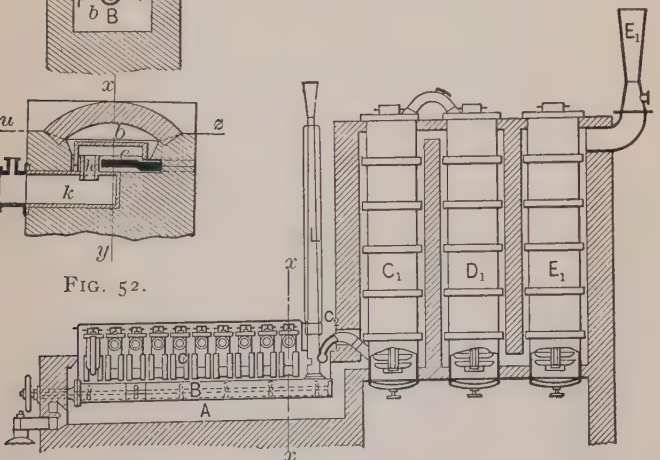

FIG. 53.

salt, have the form of iron troughs, and are heated by flame-gases (*A*); in each of them a series of cells is suspended, in which the carbon anodes and the metal cathodes are placed, which latter consist for the most part of aluminum, and are separated from

the anodes by the porcelain cylinders G. In the electrolysis the aluminum is separated at the cathode, while the chlorine developing at the anode, mixed with the vapor of chloride of aluminum, escapes through the tube g, in order first to stream into the columnar apparatus C_1D_1. The latter consists of a number of compartments, which are charged with a composition of alumina and dry, large-grained carbon. The chlorine changes the alumina into chloride, which, brought back into the bath B, there, with the excessive sodium chloride, rebuilds the double salt. The third column, E_1, is merely for the purpose of previously drying the alumina and carbon intended for the other two columns, C_1 and D_1. The heating of the columnar apparatus is effected by fire-gases, whose current is regulated by means of a steam-injector. The screw-mover b provides for the thorough mixing of the melt during the electrolysis.

Dhiel Process.—Alum, sodium fluoride, calcium chloride or magnesium chloride, and sodium sulphate are mixed in a sufficient quantity to obtain, through a double transposition, aluminum-sodium double fluoride and alkali sulphates, which latter are separated by washing. The fluoride is melted with sodium chloride and fluor-spar, and the molten mass subjected to the electrolysis.

Fig. 54 represents the crucible, as used especially by Dhiel. The anode F consists of carbon;

if one desires to obtain pure aluminum, carbon is also employed for the cathode; otherwise copper or iron is used, according to the alloy which is

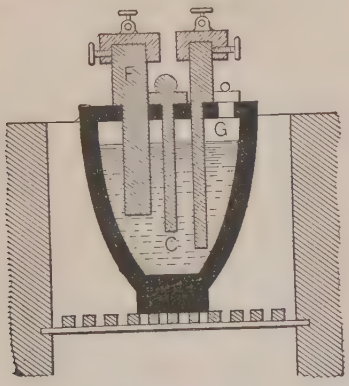

FIG. 54.

to be produced. *c* is a partition which separates the two electrodes.

Douglas-Dixon Process. — This patent is exactly like Bull's, with the single difference that magnesium is employed as reducing-metal. A composition of 35 parts $MgCl_2$, 25 parts KCl, 40 parts NaCl, mixed with 3–5% aluminum-sodium double fluoride, is electrolyzed. Before the electrolysis the mixture is heated in a melting-crucible to about 800° C.

The tension of the electrodes amounts to 7–8 volts. $MgCl_2$ is decomposed into magnesium, which rises to the surface of the melt, and into chlorine, which escapes through openings let into the top of the crucible and streams into a retort in which is found

a mixture of alumina, carbon, and common salt. The alumina is dissolved by chlorine with the formation of aluminum chloride, which unites with sodium chloride to form a double salt. The reaction is expressed in the equation

$$3C + 6Cl + Al_2O_3 + 6NaCl = Al_2Cl_6.6NaCl + 3CO.$$

The temperature is kept sufficiently high to melt the chloride without volatilizing it. When it has arrived at this state in the crucible, it is reduced by the magnesium floating upon the surface of the bath, according to the equation

$$Al_2Cl_6.6NaCl + 3Mg = 2Al + 3MgCl_2 + 6NaCl$$

with a simultaneous regeneration of magnesium chloride, which remains in the melt. In Fig. 55 diagram *a* shows the apparatus as employed in the process. The graphite crucible *A* stands upon a grate directly under the retort *B*. This communicates with a condenser, which takes up the carbonic oxide and the volatile chloride developing.

A modified arrangement is seen in *b*. While in the former instance the crucible was at the same time the cathode, here such is not the case. The pin *a* is here the negative electrode. The retort stands at the side of the crucible, and is united with it by means of the tube *a'*.

At *c* the mixture of alumina and carbon is separated from the reducing-bath only by the porous partition-wall *m*, so that the aluminum chloride,

which is formed by the chlorine developed at the anode, is directly reduced at the surface of the bath.

The process of reduction by means of the device *d* is essentially different from the foregoing. The

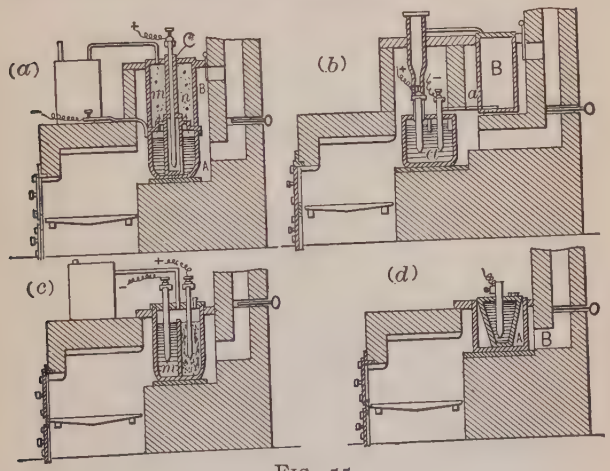

FIG. 55.

crucible *A* contains the charge, which consists of 95 parts magnesium chloride, 75 parts chloride of potassium, and 6–7% fluor-spar. Retort *B* contains the alumina, which is reduced by means of the magnesium separated at the crucible wall (the crucible being here again the cathode):

$$3Mg + Al_2O_3 = Al_2 + 3MgO.$$

The aluminum is collected on the bottom of the retort *B*, while the magnesia in crucible *A* remakes magnesium chloride, which is then dissolved anew.

Process of Hampes, Kleiner.—This rests upon the electrolysis of aluminum-sodium double fluoride (cryolite) *per se*, or mingled with a salt of the alkalis or of alkaline earths. The bath is first melted in the arc, and from then on maintained in a molten-flow state by means of the current itself.

Omlot, Bottiger, and Seidler Process (Fig. 56).—Aluminum-halogen salts are melted and electrolyzed. A peculiar feature of this patent, according

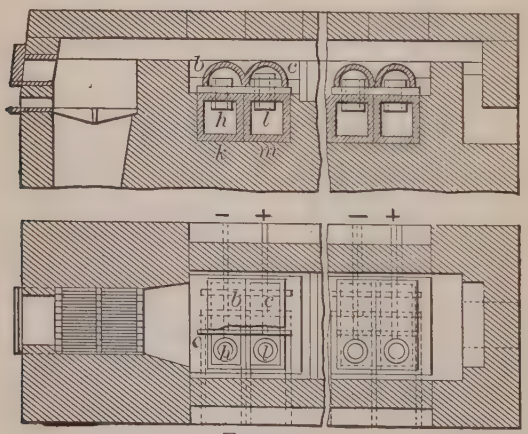

FIG. 56.

to which, it is said, operations are carried on in Crossnitz, consists in the employment of muffles *b*, *c* without floors, which are immersed in the melt and of which one contains the positive, the other the negative electrode; both electrodes alike are deeply immersed in the bath, which by means of an exterior source of heat is kept molten.

The halogens escape through the opening *l*. The

muffles consist of fire-clay with carbon lining, and are cemented air-tight; an indifferent gas provides for the exclusion of air.

Roger Process.—In the course of his researches with reference to the production of aluminum, Roger is said to have been led to mix with the aluminum salt an alloy of lead and sodium; this operation, according to the inventors' statement, should materially increase the output.

The lead-sodium alloy will receive, by electrolysis, a common-salt melt, with molten lead for the cathode.

Lossier Process.—This process depends upon the electrolysis of aluminum fluoride, which is formed chemically in the melt. Lossier, with this end in view, introduces into the melt a quantity of calcium fluoride and aluminum silicate ($Al_2O_3.SiO_2$), which is converted at the prevailing temperature into aluminum fluoride, which is to be electrolyzed, and into calcium silicate, which remains floating in the bath:

$$3CaF_2 + Al_2O_3.3SiO_2 = Al_2F_6 + 3CaO.SiO_2.$$

The metal obtained includes considerable quantities of silicon. Since under the prevailing conditions the density of the melt is greater than that of the aluminum, the latter does not sink beneath, but rises to the surface of the electrolyte; and a great loss of metal is hereby suffered, since the

metal cannot be assembled quickly enough to prevent it from being oxidized.

Bucherer Process* and the Aluminium-Industrie-Aktien-Gesellschaft Process.† — Both patents, which date from the same year (1890), rest upon the separation of aluminum from a molten solution of aluminum sulphide in chlorine alkalis. According to Bucherer, alumina may be changed in two ways into the aluminum-sodium double sulphide: either by treating alumina by heating with sodium sulphide, carbon, and sulphur:

$$3Na_2S + Al_2O_3 + 3C + 3S = Na_6Al_2S_6 + 3CO,$$

or at white heat, through the effect of carbon and sulphur upon the oxide:‡

$$Al_2O_3 + 3C + 3S = 3CO + Al_2S_3.$$

The sulphides thus obtained are dissolved in molten alkaline chlorides and subjected to electrolysis. If the necessary heat of reaction is furnished by means of the electrical current alone, the tension amounts to 5 volts; if, on the other hand, a portion of the heat is added from outside, from 2.3 to 3 volts is sufficient.

Péniakoff Process and Gooch Process.—Here, likewise, we have to do with the electrical decomposition of aluminum sulphide. With regard to the Péniakoff process for aluminum production we pos-

* D. R. P. No. 63995, Nov. 18, 1890.
† D R. P. No. 69909, Nov. 18, 1890.
‡ Zeitschrift für angewandte Chemie, 1892.

sess, it is true, only very scanty data, so that we are obliged to content ourselves with a mere reference. Much better known and also of more recent date is the process of Gooch, which rests upon the electrolysis of aluminum sulphide, the latter being produced in the electrolytic bath at the expense of the alumina dissolved therein. The inventor

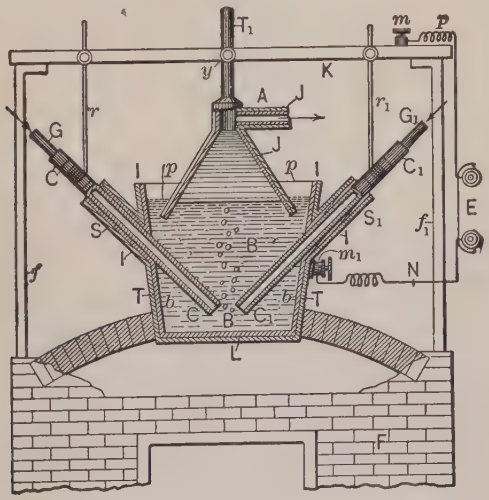

FIG. 57.

mixes a composition of sodium fluoride and aluminum chloride. He completes the melt with the addition of alumina, and conducts a current of bisulphide of carbon through it. This, according to Gooch, is produced in the electrolytes directly before the introduction, by conducting sulphur-vapors over a thick layer of carbon brought to a red glow; the precaution must be taken, however,

to generate the bisulphide of carbon beforehand, independently of the electrolysis. Furthermore, Gooch believes that the bisulphide of carbon may be replaced by any other sulphur compound—for example, by sulphuretted hydrogen.

The alumina dissolved in the bath is changed by the gas into the sulphide, and the latter is immediately decomposed by the electric current, with the separation of aluminum.

The apparatus described by the inventor in his English patent (No. 16555 of August 15, 1899) is depicted in Fig. 57.

T is an iron crucible, whose bottom and walls are lined to the height of the anodes with a layer of carbon. The walls above the anodes and the tubes *SS'*, in which the anodes *CC'* slide, are lined with alumina, so that the anodes are introduced insulated into the crucible. The anodes are connected with the positive pole of the dynamo by means of the pins *rr'*, the bar *K*, and the cable *P*. Clamp *m'* and cable *N* are the means of uniting the crucible to the negative pole of the machine.

The anodes are hollow and provided with tube feeders *GG'*, which serve for the introduction of bisulphide of carbon. The cap *J*, above the crucible, consists of iron, lined with carbon. *R* is a drawing-off tube. This chimney-like headpiece *J* is supported by the beam *T'*, which may be fastened upon the bar *K* by means of the pressure-screw *y*. During the electrolysis the cap is dipped

slightly into the melt, whose upper surface, remaining free, is covered with a carbon layer *p*.

When the apparatus is to be operated, first of all a composition of aluminum chloride and sodium fluoride is melted down in the crucible, then alumina is added and bisulphide of carbon introduced. The effect of the latter, as we have already remarked above, is to make aluminum sulphide from alumina; the aluminum sulphide is dissolved in the melt, also carbonic oxide and carbonic oxysulphide, which escape through the drawing-off tube *R*.

The sulphide is decomposed by the electric current; at the walls of the crucible aluminum is separated, which is assembled at the bottom of the crucible, while sulphur escapes. The regeneration of the bisulphide of carbon follows, according to a peculiar process described in the patent.

The inventor asserts, furthermore, that he has obtained very good results with the electrolysis of a compound of aluminum fluoride and alkali fluoride, with the addition of alumina and the introduction of bisulphide of carbon.—The Gooch process does not appear as yet to have been technically utilized.

PART II.

ALUMINUM AND ITS ALLOYS. METHODS OF WORKING AND USES.

A. The Aluminum Industry.

Since the year 1889—despite numerous assertions to the contrary—there has been a very remarkable increase in the use of aluminum in commerce and in industry; the metal is at present utilized in all forms and dimensions, from thimbles, visiting-cards, etc., which weigh but a fraction of a gram, to objects of several tons' weight, such as ship-propellers and the like.

Among the metals with which aluminum is alloyed the most important are iron, copper, nickel, and German silver. The forms in which the metal is utilized, either by itself or in alloy, are exceedingly numerous; in commerce we recognize bar-, wire-, plate-, tube-, and powdered aluminum.

The percentage of iron should not exceed 2%; from 3 to 6% of other metals may be present. The alloys possess a tensile strength of 25–35 kg per mm^2, with an elongation of 5–10%; the pure metal, on the other hand, annealed, shows a tensile

strength of only 15–20 kg per mm^2, with an extension of 30–50%.

Aluminum may also be used for heavy alloys. Copper or brass alloyed with 3–10% of aluminum gives bronzes capable of a high resistance.

Aluminum has been the subject of a large number of researches, which have had reference in part to its chemical constitution (pure metal or alloy), in part to the method of working the metal, its resistance to chemical influences (sea-water, atmosphere), its analysis, and its metallurgical and chemical utilization as a reducing-agent.

Production of Aluminum. — It is doubtless to the electrolytic methods that aluminum owes its increasing production and consumption of late years. The world's production of aluminum, which even in the year 1889—hence at a time when the electrolytic production of aluminum was just beginning its rapid development—did not amount to more than 70 tons, increased in the year 1900 to 5000–6000 tons.

The buying-price of aluminum, during the same period, fell from 30 to 3 fr. per kilogram of the metal. In the year 1855, at the time of Deville's researches, the kilogram of aluminum cost 1000 fr.; in the succeeding year the price fell to 375 fr. per kilogram.

Morin in Nanterre (1857) lowered the price to 280 fr.; from 1857 to 1886 Merle & Co. and later Pechiney, in Salindres, kept it at about 125 fr. From 1886 to 1892 England operated with the

chemical processes of Netto and Castner, which represent the perfection of Deville's method, but which were not able to furnish aluminum at a price lower than 20 fr. per kilogram. Carefully planned as these processes were, they finally had to give way to the electrolytic methods, which lowered the price of aluminum to 3 fr., and thus converted the metal into one industrially available.

Table IX shows the increase in the production of aluminum in the various countries, from 1885 down to our own day. (See Appendix, page 218.)

TABLE IX.

PRODUCTION OF ALUMINUM.

(In tonnes = 1000 kg.)

Year.	U.S.A.	Switzerland	France.	England.	Germany.
1885	1	....	2	1	10
1886	2	....	3	1	10
1887	8	....	2	1	15
1888	8	....	4	11	15
1889	22	....	15	34	15
1890	28	41	37	70	
1891	76	169	36	52	
1892	134	237	75	41	
1893	141	437	137		
1894	370	600	270		
1895	417	650	360		
1896	590	700	500		
1897	1184	800	500	300	
1898	1300	960	600	360	
1899	1500	1120	700	420	300
1900	1650	1232	800	500	500
Total production	7431	6946	4041	1791	850

In respect to the amount of aluminum produced, the United States is in the first rank; then follow Switzerland, France, England, and Germany. Thanks to the new establishment in Rheinfelden, which was erected by the Aluminium-Industrie-Aktien-Gesellschaft, the production of Germany will soon be equal to that of France and Switzerland; indeed, this may already be the case to-day.

Of all countries producing aluminum, France has decidedly the most favorable local conditions, for it is a country possessing not merely natural sources of power, which permit of the easy and successful enlargement of the new industry, but also—in contrast to other countries—extensive deposits of bauxite, which furnishes the necessary originative material for the production of aluminum.

According to Table IX, the total production of aluminum from 1885 to and including 1900 amounts to 21060 tonnes. The 5000 tonnes which were produced in the year 1900 represent a purchase-price of $13\frac{1}{2}$ million francs, and demand an electrical power of 25000 h.p., employed uninterruptedly night and day. The available energy which would be at the disposal of all the establishments which produce aluminum, when worked to the extent of their capacity, is far greater; it amounts to 61000 h.p.

The capital invested in the aluminum industry is very considerable. The capital of the Compagnie

Establishment.	Place.	Process.	Available Energy, H.P.
Compagnies des produits chimiques d'Alais et de la Camargue	*France:* St. Michel (Savoy)	Minet-Hall	6000
Société électrométallurgique française	La Praz (Savoy), Froges (Isère), Gardannes (Bouches du Rhône)	Héroult	6000
Aluminium-Industrie-Aktien-Gesellschaft	*Switzerland:* Neuhausen	Héroult	6000
	Germany: Reinfelden	Héroult	5000
	Austria: Land-Gastein	Héroult	4000
Pittsburg Reduction Company	*United States:* Niagara Falls	Hall	20000
British Aluminium	*England:* Foyers (Scotland)	Héroult	14000

des produits chimiques d'Alais et de la Camargue, invested in the factory at St. Michel, amounts to 2.2 million fr., that of the Pittsburg Reduction Company to 15 million fr.; the Société électrométallurgique française has a capital of 10 million fr.; the aluminum establishments of Switzerland represent a property of 18 millions, the British aluminium 15 millions, the Aluminum-Industrie-Aktien-Gesellschaft 19 millions; the total capital invested, therefore, amounts in round numbers to 79 million francs.

The majority of these companies unite with the industrial production of aluminum still other branches of production: the electrometallurgical

production of sodium and magnesium, the production of ferro-silicon, ferro-chrome, ferro-manganese, of metallic carbides and metallic silicides, etc.

Cost of Producing Aluminum.—An exact statement as to the cost of a kilogram of aluminum is, in view of the large number of elements entering into the question, hardly possible; the cost is, of course, influenced by the greatest variety of circumstances: by the electromotive force, the rate of compensation for labor, the electrode material, the originative material (alumina and natural or artificial cryolite), the labor of refining, etc. Nevertheless we give below some figures which obtain for an establishment of 1000 horse-power, working with water-power (average head 100–150 m).

Electric Energy.—In general it may be said that the production of a kilogram of aluminum requires in round numbers 40 electric horse-power hours. Under the specified conditions (water-power) the effective horse-power hour, measured at the electrolytic apparatus, costs 1 centime, including general oversight, surveillance of the flow of water, of the turbines and the electrical machines. The kilogram of aluminum, then, in so far as the electrical energy is concerned, comes to 0.40 fr.

Electrodes.—With apparatus in which the crucible serves simultaneously as cathode, the costs of the cathode material may be reckoned at 200 fr. for

800 kg aluminum, which makes 0.25 fr. per kilogram of aluminum produced. On the other hand the waste of the anodes amounts to about 1200 g per kilogram of the metal; since the kilogram of carbon anode may be taken at 0.20 fr., we have to allow for every kilogram of aluminum an anode waste of 0.24 fr.; taken altogether, then, a kilogram of aluminum costs 0.49 fr. in electrode material.

Payment for Labor.—For the supervision of a group of apparatus which produces 50 kg aluminum one laborer suffices for day and night service. This represents an expenditure of 10 fr. per 50 kg, hence 0.20 fr. per kilogram of the metal.

Originative Materials.—Under this classification belongs the charging of the bath at the beginning of the electrolytic process with, on the average, 50% of cryolite and 50% of chlorides and fluorides of the alkalis and alkaline earths; also the maintenance of the process, and the completion of the melt with anhydrous alumina during the course of the electrolysis. As for the original charge of the bath, we must calculate the requisites for this as costing on the average 0.30 fr. per kilogram of aluminum. The continued filling up demands 2.2 kg of anhydrous, chemically pure alumina, the kilogram costing 0.50 fr.; consequently the cost is 1.10 fr. per kilogram of the metal; the total cost is, therefore, 1.40 fr. To this should be added the expenditure for the remelting, which amounts to about 0.10 fr.; so that we may reckon the costs

of material for a kilogram of metal at a total of 1.50 fr.

Maintenance of the Establishment; Unforeseen Expenditures.—In the price of 1 centime for the electric horse-power hour we have included merely the paying off of the capitalization. For the maintenance of the establishment (melting-furnaces, workshops, chemical products, etc.) we may name the sum of 15000 fr. for a yearly production of 150 tons of aluminum; that is to say, 0.10 fr. per kilogram of metal; the unforeseen expenditures may be reckoned at about the same amount.

To recapitulate what we have said, the cost of manufacture may be stated as follows:

Cost of Manufacture for a Kilogram of Aluminum.

		Francs.
Electrical energy: 40 electric horse-power hours		0.40
Electrodes	Cathode: 200 fr. for 800 kg aluminum = 0.25 fr.	
	Anode: 1200 g @ 0.20 fr. per kg of anode-weight = 0.24 fr.	0.49
Cost of labor: two laborers @ 10 fr. for 50 kg aluminum		0.20
Materials:	Charge and completion of the bath. 0.30 fr.	
	Alumina 22. kg @ 0.50 fr. 1.10 fr.	
	Remelting (coke and crucibles) 0.10 fr.	1.50
Maintenance and unforeseen expenses		0.20
Total		2.79

We thus obtain a price in the neighborhood of 3 fr., which, in fact, is the actual purchase-price. To be sure, most establishments avail themselves of a greater power than we have assumed in the above reckoning, so that they can obtain the horse-power

hour more cheaply; furthermore, we must not overlook various improvements which have been made, as for example in the production of the electrodes and of the originative materials, especially alumina. Further investigations will undoubtedly lead to yet further improvements; however, it may certainly be said that the electrometallurgy of aluminum has already attained a degree of perfection beyond which we can scarcely make any material advance.

B. Aluminum and its Alloys.

Aluminum is employed not merely in the pure condition, but also as an alloy, namely, as a constituent of light, heavy, and medium-weight alloys.

(*a*) Pure Aluminum.

Pure aluminum is used where the requirement is not extraordinary mechanical stability, but strong resistance to chemical influences.

The atomic weight of aluminum is 27.08. Its density varies, according to the treatment to which it is subjected, between 2.6 and 2.74. It melts at 650° C. and has a white color and, especially on freshly cut surfaces, a beautiful lustre. In the air it does not change appreciably if it is free from silicon. If it contains this element to a considerable extent (0.5–1%), an exchange would appear to take place in the interior of the metal; the silicon goes to the surface, is oxidized, and forms a

thin layer of silica, which may be wiped off at a touch.

Aluminum is able to reduce almost all oxides, even those of carbon, silicon, and boron. Water and dilute organic acids scarcely affect aluminum at all; at a boiling heat, it is true, it is attacked by organic salts, but only very gradually. Nitric acid is almost entirely ineffectual; by sulphuric acid aluminum is dissolved gradually, by hydrochloric acid and by alkalies rapidly and easily. Below, in the part which treats of the working of aluminum, we shall adduce some further researches, which have to do with its resistance to chemical reagents.

Mechanical Properties and Electrical Conductivity.—Charpentier-Page of Valdoie (Belfort District) has instituted some very interesting experiments with regard to the mechanical properties of pure aluminum and of its alloys, as well as their electrical conductivity. We give below some of his results; in the first place, those that have to do with pure aluminum in the form of wire.

TABLE X.

Pure Aluminum.

ANNEALED WIRE.

2 mm. diam. Density 2.688.

Electrical Tests.

Resistance per metre	0.00919Ω
Resistance of a wire of 1 mm² cross-section per km.	28.86Ω
Resistance of a copper wire of the same dimensions, at 22° C.	17.9Ω
Proportion of conductivities	62%

Mechanical Tests.

	Test.		
	1	2	3
Length of the test-piece. ....mm	110	110	110
Elongation. ..... "	36	35	34.5
Elasticity. ..... kg	32.70	33.2	33.2
" per mm². ..... "	10.5	10.57	10.57
Elongation. ..... %	32.7	31.8	31.3

HARD WIRE.

2 mm diam. Density 2.694

Electrical Tests.

Resistance per metre. 0.00928Ω
Resistance of a wire of 1 mm² cross-section per km. 29.15Ω
Resistance of a copper wire of the same dimensions, at 22° C. 17.9Ω
Proportion of conductivities. 61%

Mechanical Tests.

	Test.		
	1	2	3
Length of the test-piece. ....mm	110	110	110
Elongation ..... "	4.5	4.5	4
Elasticity. ..... kg	72	72.7	72.5
" per mm². ..... "	22.9	23.14	23.05
Elongation. ..... %	4	4	3.6

From this tabulation may be seen the advantage aluminum offers as conducting material. If one compares the above figures for aluminum with those for copper, one sees that at current prices for both sorts of wire aluminum wire with the like conductivity is cheaper than copper wire. If 100 is the conductivity for copper, 62 is that

for aluminum. 8.95 is the density of copper, 2.67 the density of aluminum; a kilogram of copper wire costs 2.75 fr., a kilogram of aluminum wire 3.75 fr. With a like conductivity, the cross-section of the aluminum wire $S_c = \frac{100}{62} = 1.61$, that of the copper wire S_c being placed equal to 1. Under the same conditions the weight of the aluminum wire $Q_a = 1.61 \times 2.67 = 4.3$, that of the copper wire $Q_c = 1 \times 8.95 = 8.95$. The price, therefore, for aluminum wire stands at $P_a = 4.3 \times 3.75 = 16.13$ fr.; for copper wire, on the other hand, at $P_c = 8.95 \times 2.75 = 24.6$ fr.

Uses.—Aluminum in the pure state serves for the manufacture of electrical conductors, for surgical apparatus, precision-instruments, artistic objects, cooking utensils; also in chemistry as a reducing-agent in the production of certain metals, such as chrome, manganese, vanadium, uranium, etc.

(*b*) Heavy Alloys.

Among the heavy alloys are reckoned the various kinds of aluminum bronze and brass, and also some alloys in which zinc predominates, such as those of Cothias. The first-named aluminum alloys have already been utilized industrially for a long time, because of their notable mechanical properties. They take a high polish and withstand atmospheric influences excellently.

Aluminum Bronze. — Alloys with 7.5% of aluminum and 92.5% of copper have a golden color, but are less durable than similar alloys containing 10% of aluminum. In practice this proportion is seldom exceeded, since an alloy containing a higher percentage of aluminum is very brittle.

Aluminum bronzes are used, in a small way, for the manufacture of optical instruments, tableware, ornaments, etc.; on a larger scale, for ship-propellers, armor-plate, etc.

Bronzes containing 2.5, 5, and 10% correspond to the formulæ: Cu_2Al (with 9.62% Al), Cu_8Al (with 5.05% Al), $Cu_{16}Al$ (with 2.59% Al).

Aluminum Brass. — The amounts of aluminum and zinc vary. Usually the composition is as follows:

Copper.....	67	71	55.8	55.8	67.7	per cent
Zinc.....	30	27.5	42	43	26.8	"
Aluminum.....	3	1.5	4.2	1.2	5.8	

The tensile strength of the first two alloys varies from 21–45 kg per mm^2; that of the next two amounts to 50 kg; in the case of the fifth alloy a tensile strength up to 65 kg would be obtained. A proposal in *The Aluminum World* is worth noting, according to which zinc and aluminum are to be added to the copper, in the form of a previously prepared zinc-aluminum alloy containing from 5 to 10% of the last-named metal. There is a larger percentage of aluminum in the zinc-aluminum

alloys of Cothias—alloys which are easily poured and may advantageously replace cast zinc.

Researches Concerning Aluminum Bronze and Brass.—These alloys have been the subject of countless investigations, among which should be mentioned the researches of Debray, who first prepared bronzes with 10% of aluminum, then the investigations of Cowles and Héroult, who for the first time produced aluminum bronzes for technical purposes. Among the results of Héroult which relate to the bronzes obtained in Froges and in Schaffhausen, it is worthy of special note that a bronze with 10.5% of aluminum, before it has had any mechanical working, and hence in the rough state in which it has come from the melt, has an elasticity of 63.8 kg per mm^2, with an elongation of 6.8%. With a special alloy—89 parts copper, 10 parts aluminum, 1 part silicon—Cowles obtained a strength of 100.5 kg. Pouthière, professor in the University of Louvain, found in his tests in the establishment at Malines with cast bars 8 mm in diameter, which came from the Cowles works in Lockport, a strength of 69.31 kg, with a simultaneous elongation of 4.3%; the alloy in question contained 90.15% Cu, 8.10% Al, and 1.75% Si.

A noteworthy investigation of aluminum bronzes and similar alloys we owe to André Le Chatelier, who made researches into the molecular changes that occur when rolled and drawn aluminum bronzes

are heated, and compared his results with analogous conditions in the case of copper.

The following table gives the results obtained by Le Chatelier, on the one hand with steam-tubes of 10% aluminum bronze, and on the other hand with copper tubing.

TABLE XI.

ELASTICITY OF COPPER AND OF 10% ALUMINUM BRONZE, IN RELATION TO TEMPERATURE.

Temperature, $t°$ C.	Copper.		10% Aluminum Bronze.	
	Elasticity.	Elongation.	Elasticity.	Elongation.
15	25.2	30%	53.2	19%
100	22.9	30%	52.4	22%
150	20	30%	51	21%
200	16.9	30%	49.2	22%
250	14	29%	47	21%
300	12.7	20%	44.2	19%
350	9.4	15%	37	15%
400	7	10%	23.2	21%
460	3.6	....	10	23%

These figures, which need no further explanation, clearly indicate the superiority of aluminum bronze. Thus we find, for example, that at 350° C. the strength of copper has been diminished by 60%, while that of the bronze is lessened only by 40%; and that the strength of the bronze at 350° C. is practically the same as that of the copper at 15° C.

When Le Chatelier came further to compare the effect of heat on 10 and 9% cast bronze, he found that with an increase in temperature from 15° C.

to 400° C. the tenacity of the first alloy is diminished by 30%, that of the second, on the other hand, by 70%. Cast bronzes with 9 and 5% of aluminum show in the temperature-interval from 15–380° C. a like relation; in the case of both alloys the tenactiy is diminished by 30%.

If, in making bronze, the aluminum is added molten to the copper, a rise in temperature is observed; this development of heat is regarded by some investigators as proof of a chemical union of the two metals. Kiliani is nevertheless of the opinion that this increase of temperature is not conclusively and necessarily to be ascribed to such union, but in large part also to the reaction between aluminum and copper protoxide, which latter is always contained in commercial copper. He advances the following reason for this hypothesis: If one part of aluminum be added to nine parts of copper,—not all at once, however, but a little at a time,—there is presently a significant increase of temperature, while the last portions of aluminum, owing to the latent heat of fusion, effect a lowering of the temperature.

Aluminum bronzes strongly resist salt solutions and sulphurous liquids. In the laboratory of the works at Neuhausen plates of various alloys were exposed for fourteen days to the action of solutions which contained 3% of cooking-salt and 4% of acetic acid. The relative losses in weight were as given below:

	Loss in Weight.
Bronze with 10% aluminum, free from silicon	1 part
" " " " with 2.8% Si	2.1 parts
Brass " 3.5% aluminum	4.4 "
Phosphor-bronze	32 "

The same alloys, on being exposed to sea-water, showed losses in weight as follows:

	Loss in Weight.
Bronze with 10% aluminum, free from silicon	1 part
" " " " with 2.8% Si	39 parts
Brass with 3.5% aluminum	101 "
Phosphor-bronze	116 "

In the *Journal of the Society of Chemical Industry* are assembled a great number of results, based upon manifold tests, which concern themselves with various sorts of steel, with pig iron, cast iron, cannon bronze and aluminum bronze. The most important data are reproduced herewith:

TABLE XII.

Steel, Pig Iron, Cast Iron.

	Elasticity per mm^2 Cross-section.
Cannon-steel, hardened, annealed and rolled	69.70 kg
Steel, neither hardened nor annealed	63.80 "
" , cast	51.30 "
Puddled iron, melted; in thin bars	52.90 "
" " " medium thick	38.40 "
Forged iron	34.90 "
Cast iron	21.70 "
Firminy steel for the French artillery	71.20 "

Cannon Bronze.

Copper 88, Tin 10, Zinc 2 parts	28.30 kg
" 92, " 8, " 2 "	21.10 "
" 91.7, " 8.3, " 2 "	22.50 "

Aluminum Bronze.

Copper 89,	Aluminum 10,	Silicon 1 part.	76.10 kg
" 91.50	" 7.5	" 0.75 parts. .	50.60 "
The same, cast.	47.10 "
" " , rolled.	60.20 "
Copper 95,	Aluminum 5	parts, rolled.	60.10 "
" 92.5	" 7.5	" "	44 "
" 91	" 9	" "	55.90 "
" 90	" 10	" "	69.80 "

Waldo, dissenting from Kiliani's conclusion, believes that he has demonstrated that aluminum bronze is not a simple alloy of copper and aluminum, such as, perhaps, the alloy of copper and tin (with the exception of the compound $SnCu_3$), but a perfectly definite chemical compound. Waldo advances several reasons in support of this hypothesis, among others the considerable quantity of heat which is freed when one mixes the two metals, aluminum and copper, in a molten state; Kiliani's objection to this argument we have discussed above.

Likewise from the dependency of the electrical conductivity of copper upon the amount of aluminum contained therein, Waldo concludes that we have here a chemical compound, and no ordinary alloy. One actually sees, from the respective curves of conductivity, that the electrical behavior of the alloy is dependent in an unusual degree upon the proportion of aluminum; the curve shows that the addition of even the minutest quantity of aluminum exerts an influence disproportionately great upon the conductivity,—a circumstance which goes to support Waldo's contention. Furthermore, Waldo

refers to the fact that we are unable to discover any simple method of separating aluminum from copper when we have both metals combined.

If one takes a large piece of 10% aluminum bronze, it is possible to discern neither the trace of any natural joint between the two metals, nor yet the presence of grains of aluminum in the mass of copper, nor any other differences of constitution in the alloy.

Tests quite analogous to those instituted by Charpentier-Page for pure aluminum have been carried out by the same observer on 10% aluminum bronze.

TABLE XIII.

Aluminum Bronze.

Aluminum 10, Copper 90 parts.

Hard Wire, 1.6 mm in diam. Thickness 8.2.

Electrical Tests.

Resistance per metre	0.668Ω
Resistance of a wire of 1 mm² cross-section per km	133.60Ω
Test temperature	18° C.
Resistance of a copper wire under conditions otherwise similar	17.5Ω
Proportion of conductivities	13.15%

Mechanical Tests.

	I.	II.
Length of test-piece in metres	0.10	0.10
Length at the load-limit reached	0.130	0.128
Elasticity kg	129	128
Elasticity per mm² cross-section kg	64.5	61
Elongation mm	30	28

We see from these figures that a practical application of aluminum bronze for conducting material is not to be considered.

(*c*) Alloys of Medium Density.

In this group belong only a few alloys which are used industrially as metals, many, however, which serve for solder; we shall reserve the detailed description of the latter for the section which treats of the "Working of Aluminum." Here we shall mention only colored alloys with gold, palladium, cobalt, and nickel; also a special iron-silicon-aluminum, which is of importance in metallurgy.

Aluminum-gold Alloy.—This was produced for the first time by the English chemist Roberts-Austen; it contains 22 parts aluminum and 78 parts gold, and is of a purple color, with a ruby lustre.

This alloy appeared for a while destined to play a rôle for ornamental purposes, and as a coin-metal; but after the researches of Margot it was impossible to deny that the metal was practically useless, since it possesses neither the ductility nor the malleability to make it possible to work or stamp the metal for such purposes.

The brothers Tissier found that aluminum could be alloyed with gold up to 10% without losing its malleability. The so-called "Nürnberger gold" is an aluminum alloy well suited for artistic objects, has a golden color, and strongly resists atmospheric influences. Its composition is as follows: 90 parts

copper, 2.5 parts gold, and 7.5 parts aluminum. On the basis of its specific weight it belongs rather in the category of heavy alloys.

Aluminum-platinum Alloy.—This was first produced by Margot, assistant at the University of Geneva; it contains 28 parts aluminum and 72 parts platinum, has a beautiful yellow color which, with certain slight variations on account of its chemical composition, takes on a vivid greenish and sometimes copper-like lustre, is brittle, hard, and of crystalline structure.

Aluminum-palladium Alloy (Margot).—It consists of aluminum and palladium in proportions such as those of the preceding alloy, has a beautiful rose color which, as soon as its composition is slightly altered, passes over into steel-gray, is likewise of crystalline structure, brittle, very fragile, without, however, having the tendency to crumble gradually.

Aluminum-cobalt Alloy (Margot).—It contains aluminum to 20–25% and cobalt to 75–80%. Freshly produced, it is as hard as hardened steel, crystalline, and, like the alloys just described, crumbles away completely upon being hammered. Even after a few days it falls into a powder of a pronounced violet-blue tint.

Aluminum-nickel Alloy (Margot).—18% aluminum, 82% nickel; of a clear straw-yellow color; almost as hard as steel and capable of taking a high polish. In contrast to the previous alloys it may be

hammered without altering its constitution in any way.

Ferro-silicon-aluminum (Minet).—Under this classification belong alloys having the following composition:

Aluminum.	Iron.	Silicon.
90	7	3 parts
85	10	5 "
80	14	6 "

They are produced directly in the electric furnace from white or red bauxite or from a mixture of the two, and are successfully used in metallurgy.

(*d*) Alloys of Various Densities.

Among the most recent investigations of the aluminum alloys we shall mention the researches of Leon Guillet on the alloys with wolfram and molybdenum, then the researches of Boudouard on magnesium-aluminum, and those of Edmond Van Aubel on antimony-aluminum.

Researches of L. Guillet.*—This investigator has availed himself of the Goldschmidt aluminothermic process for the production of his alloys. His earliest investigations relate to the reduction of tungstic acid, molybdic acid, magnetic iron ore, manganese monoxide, and titanic acid by means of aluminum in excess.

* L'Electrochimie, June 1901, pp. 86 and 89; July, p. 119.

We shall first describe the results which Guillet has obtained in the reduction of tungstic acid.* Neither tungsten nor aluminum must be present in too great excess: tungsten must not be present in excess, or the reaction will be too active; the presence of aluminum in excess would prevent the compound from being enkindled. A mixture which yields $Al_{10}W$ as the reaction-product stands just on the border-line of inflammability.

Tests in which the composition of the original material is so selected that they lead theoretically to alloys of the formulas AlW_{10} and Al_5W give a metal regulus which, treated with nitro-hydrochloric acid, leaves behind a beautifully crystallized residuum with the composition AlW_2 (W 93.16%, Al 6.84%). The crystals are readily affected by concentrated acids, and dissolved by boiling water.

Tests in which, theoretically, alloys AlW and $Al_{10}W$ should be obtained yield numerous laminated crystals of the formula Al_4W (W 63.02%, Al 36.98%), which are likewise affected by concentrated acids.

Tests which, theoretically, should yield alloys with the composition Al_3W and AlW_5 give crystals which at the surface of the metallic mass form beautiful excrescences, and for which we have the formula Al_3W (W 69.34%, Al 30.66%). These crystals are but slightly affected even by

* Compt. rend. de l'Acad. des sciences, May 6, 1901, Paris.

concentrated acids; they disintegrate, however as, do both of the other alloys, in boiling water.

Besides tungsten-aluminum L. Guillet succeeded in producing molybdenum-aluminum * as well, with the aid of the alumino-thermic process: six compounds, indeed, with the formulæ Al_7Mo, Al_3Mo, Al_2Mo, AlMo, $AlMo_4$, and finally an alloy very rich in molybdenum, which seems to have the composition $AlMo_{20}$.

Researches of Boudouard † on Magnesium-aluminum Alloys.‡—In the year 1866 Wöhler began to attempt the production of alloys of aluminum and magnesium, by fusing the two metals together with common salt.§ He obtained in this way a mixture which produced a lustrous tin-white powder, but without any perceptible crystallization.

Later Parkinson‖ succeeded in obtaining a product with 25% of magnesium, by melting down both metals in a crucible charged with pure, fresh magnesia. As for the effect of the magnesium upon the properties of the alloy in question, we may say, in general, that a percentage of magnesium makes any alloy brittle and liable to crumble.

Very lately Mach has produced an aluminum alloy with 10–12% of Mg, which in consequence of

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* Compt. rend. de l'Acad. des sciences, June 3, 1901, Paris.
† L'Electrochimie, June 1901, p. 88.
‡ Compt. rend. de l'Acad. des sciences, June 3, 1901, Paris.
§ Annal Ch. Pharm., CXXXIII, 253.
‖ Chemical Society (2), Vol. V, p. 117.

the presence of the latter is lighter than pure aluminum, is of a silver hue, and may be worked in any way desired.

Boudouard, who made it his special object to determine the melting-point of the different aluminum-magnesium alloys, obtained the following results:

TABLE XIV.

MELTING-TEMPERATURE OF THE ALUMINUM-MAGNESIUM ALLOYS.

Aluminum, Per Cent.	Magnesium, Per Cent.	Melting-point, Degrees C.
100	0	650
90	10	585
80	20	530
70	30	432
60	40	450
50	50	462
45	55	445
40	60	450
35	65	455
30	70	424
25	75	356
20	80	432
15	85	432
10	90	437.5
5	95	595
0	100	635

If a curve is constructed with weight-percentages of aluminum for abscissæ and the melting-points as ordinates, it is seen that this curve has two maxima at 455° and 462° C., and three minima at 356°, 445°, and 432° C. Between 10% and 20% aluminum the curve is clearly parallel with the axis

of the abscissæ. The two maxima express the two well-defined compounds $AlMg_2$ and AlMg.

As for the malleability, only alloys with a percentage of aluminum or magnesium not higher than 0–15% can be considered. An alloy which consists half of aluminum, half of magnesium, crumbles to pieces in the fingers, and may be powdered in porcelain mortars.

Researches of E. van Aubel on Aluminum-antimony Alloys.*—An alloy whose composition is expressed in the formula AlSb melts at 1078–1080° C., while the pure metals melt at 660° and 430° C. respectively. Aubel has investigated the question whether the formation of this peculiar alloy is connected with an alteration in volume. Tests in which two pieces with known percentages of aluminum and antimony were taken at different places gave a complete homogeneity and a percentage composition of 18.87% Al and 81.13% Sb. The density of this alloy, referred to the vacuum and to water at 4° C., amounts to 4.2176 at a temperature of 16° C. It is, therefore, considerably smaller than we might theoretically expect, and it follows that with the formation of the alloy a very considerable increase in volume takes place. We have here, then, an exception to the Matthiesen law. We may also formulate the result more clearly,—that 7.07 cm^3 aluminum + 12.07 cm^2 antimony give 23.71 cm^3 of the alloy AlSb.

* L'Electrochimie, September 1901, p. 136.

(*e*) Light Alloys.

Under this heading belong a great number of alloys, which are classified as "light" because their density does not differ materially from that of aluminum, since they contain, at most, 6% of their weight in heavy metals.

Copper-aluminum Alloys. — Their percentage of copper varies between 3 and 6%. Table XV reproduces the test results which were obtained by Charpentier-Page in two extreme cases.

TABLE XV.

Aluminum 97%, Copper 3.

ANNEALED WIRE.

2 mm in diam. Density 2.737.

Electrical Tests.

Resistance per metre	0.01141Ω
Resistance per mm² cross-section and per km	35.83Ω
Resistance of a copper wire of the same dimensions, at 22° C.	17.9Ω
Proportion of conductivities	49.99%

Mechanical Tests.

	Test.		
	1	2	3
Length of test-piece ..... mm	110	110	110
Elongation ..... "	23.5	23.5	25
Elasticity ..... kg	64.5	64.5	65.10
Elasticity per mm² cross-section ..... "	20.54	20.38	20.76
Elongation ..... %	21.3	21.3	21.7

HARD WIRE.

2 mm in diam. Density 2.742.

Electrical Tests.

Resistance per metre. 0.01145Ω
Resistance per mm² cross-section and per km. 35.96Ω
Resistance of a copper wire of the same dimensions, at 22° C. 17.9Ω
Proportion of conductivities. 49.77%

Mechanical Tests.

	Test.		
	1	2	3
Length of the test-piece. . . . . mm	110	110	110
Elongation. . . . . "	5	4	4.5
Elasticity. . . . . kg	110	111	109
Elasticity per mm² cross-section. . . . . "	35.3	35.3	34.7
Elongation. . . . . %	4.5	3.6	4

Aluminum 94%, Copper 6%.

ANNEALED WIRE.

2 mm in diam. Density 2.818.

Electrical Tests.

Resistance per metre. 0.01025Ω
Resistance per mm² cross-section and per km. 37.81Ω
Resistance of a copper wire of the same dimensions, at 19° C. 17.6Ω
Proportion of conductivities. 46.5%

Mechanical Tests.

	Test.		
	1	2	3
Length of test-piece. . . . . mm	105	105	105
Elongation. . . . . "	17	19	21
Elasticity. . . . . kg	78	75	73.5
Elasticity per mm² cross-section. . . . . "	24.8	23.8	22.4
Elongation . . . . . %	16.2	18	20

HARD WIRE.

2 mm in diam. Density 2.827.

Electrical Tests.

Resistance per metre	0.0129Ω
Resistance per mm² cross-section and per km	40.51Ω
Resistance of a copper wire of the same dimensions, at 19° C.	17.6Ω
Proportion of conductivities	43.44%

Mechanical Tests.

	Test. 1	Test. 2	Test. 3
Length of test-piece.....mm	105	106	105
Elongation..... "	3	2.5	3
Elasticity..... kg	142	135	135
Elasticity per mm² cross-section..... "	45.2	42.9	42.9
Elongation..... %	2.8	2.3	2.8

A simple calculation, quite like that which we have made for pure aluminum, shows the advantage of using conducting wire of copper-aluminum rather than pure copper, although the alloy in question is a somewhat poorer conductor than pure aluminum, and is only half as good as pure copper.

Copper-aluminum is in many cases preferred, in industry, to pure aluminum, because of its excellent mechanical properties.

Nickel - aluminum, Nickel - copper - aluminum, or German - silver - aluminum (Tissier, Le Verrier, and especially A. E. Hunt, technical director of the Pittsburg Co.).—The alloys named contain only about 3% of heavy metals. Nickel imparts to the

aluminum a certain stiffness, and gives an alloy which may be easily worked or made into plates, and whose mechanical properties are about the same as those of copper-aluminum.

Joseph Richards has found that, of all aluminum alloys, nickel-aluminum and nickel-copper-aluminum best withstand chemical influences. He has exposed a great number of alloys to the effect of hydrochloric acid, nitric acid, acetic acid, potash-lye, and sodium chloride, and has drawn up the following table, in which the alloys are arranged in groups according to their increase in stability.

HCl.	NHO_3.
Titanium-aluminum	Aluminum, pure
Aluminum, pure	Titanium-aluminum
Copper-aluminum	Copper-aluminum
German-silver-aluminum	Nickel-aluminum
Nickel-aluminum	German-silver-aluminum

$C_2H_4O_2$.	KOH.
Aluminum, pure	Aluminum, pure
Titanium-aluminum	Titanium-aluminum
Copper-aluminum	Copper-aluminum
German-silver-aluminum	Nickel-aluminum
Nickel-aluminum	German-silver-aluminum

NaCl.

Aluminum, pure
German-silver-aluminum
Titanium-aluminum
Copper-aluminum
Nickel-aluminum

The German silver here used is the so-called "type de la guerre," with the composition: 80% Cu, 20% Ni.

Nickel-tin-aluminum.—Of this alloy three different lots were tested:

No. 1: 85 parts aluminum, 15 parts tin, 2 parts nickel
No. 2: 90 " " , 10 " " , 3 " "
No. 3: 90 " " , 11 " " , 4 " "

These alloys he finds much harder than aluminum and easier to work with the file; moreover, they may be soldered directly with one another or with aluminum and other metals. The solder contains either 4 parts silver, 8 parts zinc, and 5 parts tin, or else 5 parts silver, 8 parts zinc, and 5 parts tin.

Nickel-iron-aluminum. — Composition: 90 parts aluminum, 4 parts nickel, 1 part iron, or 85 parts aluminum, 10 parts tin, 4 parts nickel, and 2 parts iron. These alloys may without difficulty be worked with the file, and may be easily rolled; they break in pieces, however, under the hammer.

Cobalt-aluminum. — With a proportion of 6% of cobalt this alloy may be easily rolled into plates.

Manganese-aluminum. — Michel obtains an alloy of this kind by melting together 2 parts manganese protochloride, 6 parts potassium-sodium chloride, and 4 parts aluminum. If the metal mass is treated with hydrochloric acid, an insoluble part remains, having a density 3.4, whose composition is expressed in the formula $MnAl_3$.

Manganese-copper-zinc-aluminum.—The analysis of alloys of this sort which were produced by Susini gave:

	Aluminum.	Manganese.	Copper.	Zinc.
No. 1.....	97	1–3	1.5	0.5
No. 2.....	98	1–5	2.5	1
No. 3.....	92	2–8	4.5	1.5
No. 4.....	90	10	...	...

Titanium-aluminum (Wöhler, Michel, Lévy). — Michel produced an alloy of the formula Al_3Ti, which contained, accordingly, 35% of titanium. An alloy with 70% of titanium, examined photomicrographically, gave peculiar results: it appeared as if slashed with sword-cuts. An alloy with 3% of titanium is, according to Brown, almost as hard as iron.

Tungsten-aluminum. — This alloy also originates with Michel. Its proportion of aluminum and of tungsten is expressed in the formula Al_3Wo.

The following table gives some test-results obtained by Le Verrier with an alloy containing 7.5% of tungsten:

	Elasticity per mm².	Percentage of Elongation.
Metal, cast.....	15.5 kg	1.5%
" rolled, hardened.....	25 "	4%
" annealed.....	18 "	10%
" "	15.9 "	14%

Reinhard and Isidor Roman recommend an alloy containing tungsten which they call wolframinium, and which contains 0.75 part copper, 0.105 part tin, 1.442 parts antimony, 0.0388 part tungsten,

and 98.04 parts aluminum. Its mechanical properties may be seen from the following figures:

	Elasticity per mm².	Percentage of Elongation.
Metal, hardened.....	38.7 kg	2.14%
" annealed.....	26.5 "	15.24%

Partinium.—This alloy, the name of which is derived from that of the discoverer, G. H. Partin, is obtained in the following manner: First, a mixture of 78 parts copper, 20 parts tin, and 2 parts potassium arsenate is melted; the alloy obtained is then powdered, and with it are mixed 1 part tungsten and 3 parts antimony. The whole is thereupon melted again, pulverized, and added to aluminum, which is alloyed, up to 4%, with this metallic mixture. Tungsten and antimony may here be replaced by an equal weight of powdered magnesium.

As a solder for partinium the inventor recommends a mixture of 60 parts zinc, 30 parts tin, 4 parts nickel, and 4 parts copper, which are melted with 2 parts potassium arsenate.

Zinc-aluminum.—Hard, but brittle.

Cadmium-aluminum.—Quite capable of elongation; its particular use is as a solder-metal.

Bismuth-aluminum.—With a percentage of more than 1% of bismuth, it is brittle and fragile.

Antimony-aluminum.—According to D. A. Roche aluminum is alloyed with antimony easily and in all

proportions. Alloys with a slight percentage of antimony (below 5%) are harder, more tenacious, more elastic, and at the same time more malleable than pure aluminum. Although with an increase in the proportion of antimony the hardness increases, the tenacity and the elasticity decrease very rapidly, and the alloy is readily pulverized.

Silicon-aluminum.—These alloys are always more or less ferruginous; with 1–2% of iron the strength increases with an increase in the proportion of silicon, and soon reaches 23–25 kg per mm^2 with an elongation of 10%. Unfortunately these alloys are strongly attacked in air, as well as by most chemical reagents.

Silver-aluminum. — With 5% of silver this alloy is said to be just as malleable as the pure metal. Carrol produces an alloy with 90–93 parts aluminum, 6–9 parts silver, and 1 part copper which, it is said, may be advantageously used for engravings. The addition of copper, according to the statement of the inventor, appears to give the metal a denser grain. With from 10% of silver upwards, the alloy is brittle; under the name of "tiers-argent" there has, however, been obtained an alloy of $\frac{2}{3}$ aluminum and $\frac{1}{3}$ silver, which is said to permit of being stamped and engraved more readily than copper-silver alloys.

Tin-aluminum is chiefly of consequence in connection with the manufacture of solder. Bour-

bouze recommends an alloy with 10% of tin, which is said to be soldered just as readily as brass; he employs it to advantage for physical apparatus, since its coefficient of expansion is lower than that of pure aluminum. At the same time, Riche shows that alloys of tin and aluminum are more readily attacked than either metal by itself. Its elasticity is less than that of pure aluminum, as appears from the following table:

Composition of the Metal.				Method of Working.	Elasticity per mm^2 Cross-section.	Elongation in Per Cent.
Aluminum, Per Cent.	Silicon, Per Cent.	Iron, Per Cent.	Tin, Per Cent.			
88	1.35	0.65	10	cast	9.80	4.11
48.9	0.72	0.36	50	forged	10.61	0.08

With respect to ductility also, the alloy is inferior to the unalloyed metal.

Le Verrier has made tests to determine how the melting-point of tin-aluminum varies with the percentage of tin.

Composition of the Metal.				Melting-point, Degrees C.
Aluminum, Per Cent.	Silicon, Per Cent.	Iron, Per Cent.	Tin, Per Cent.	
90	1.4	0.70	8	595
78.2	1.2	0.60	20	575
68.4	1.05	0.53	30	535
58.7	0.90	0.45	40	575
48.9	0.75	0.38	50	570
19.6	0.30	0.15	80	530
9.8	0.15	0.07	90	490

From this table it follows that the melting-point is rather independent of the composition of the alloy, at least up to a proportion of 80% of tin. With 90% of tin, however, it always remains at 490° C.

Chromium-aluminum.—Wöhler obtained an alloy of this kind by reducing the violet chromium chloride by means of aluminum; there is a metal regulus whose composition is approximately expressed by the formula AlCr. If the alloy is to be hammered and rolled, the proportion of chromium should not exceed 3%.

Quicksilver-aluminum.—According to Baille and Féry we have here a compound Al_2Hg_3. The higher the temperature with which one works, the more readily the amalgam is obtained. While at 100° the reaction is extremely sluggish, the two metals unite very quickly at the boiling-point of quicksilver. According to Krouchkoll the alloy is readily oxidizable.

C. Working of Aluminum.

We may say, in general, that aluminum may be worked as copper is worked, and with the same tools, but with more difficulty. We will, however, add just here that, despite countless experiments, no easily applicable solder for aluminum has yet been found, and also that the proposed processes for coppering, silvering, and gilding have not been conclusively tested.

As for the melting- and the casting-process, aluminum is melted dry, that is to say, with no flux, in clay or graphite crucibles, and during the melt metal is constantly added. When the mass is completely molten, it is brought to a red glow, and the crucible is removed from the fire. The metal is now violently stirred by means of an iron rod, which ends in a small, round ladle at a right angle to the rod and perforated; the surface of the melt is skimmed and the layer of oxide formed is removed, whereupon the true operation of casting begins. The stirring-rod is removed from the melt as soon as—without as yet being at a red glow—it is so hot that the metal does not adhere to it.

Since the aluminum as it stiffens shrinks quite perceptibly—about 1.8%—during the stiffening, according to the amount of contraction, molten metal should be cautiously added in as small quantities as possible, in order to keep the mold well filled. For the casting-mold metal vessels may be used; complicated objects it is advisable to cast in sand.

Pure aluminum, as well as that of commerce with 98.5% of aluminum, may be forged, drawn, and rolled cold, without necessarily being annealed beforehand. With 97% aluminum and a preparation of 3% of heavy metals and silicon it may likewise be forged and rolled cold, but only after it has been repeatedly subjected to an annealing process. It is preferable under these conditions to work the

metal heated to a temperature approximating 200° C. If the aluminum, on the other hand, contains more than 5% of foreign elements (including silicon), it can only be worked in the heat. With 1% of heavy metals present it may be rolled provided the proportion of silicon is 10–15%.

It is generally not best to heat the metal higher than to 350° or 400° C. during or at the beginning of the treatment; indeed, it may well be kept somewhat below this temperature, and heated, if necessary, higher in some parts; while the other parts, according to the mode of the further treatment, remain outside of the fire or between two iron plates, and may eventually be cooled.

As an example of the way in which aluminum may be worked by means of the wooden anvil and wooden hammer, especially in ship-building, we may cite the pleasure-yacht "Vendenesse," belonging to Count J. de Chabannes la Palice, in whose bottom and rudder-post aluminum was used extensively. This vessel was built by Godinot after the plans of Victor Guilloux; for metal a 3% copper-aluminum alloy was employed, which was forged and worked cold, and was found to answer very well. The thickness of the aluminum plates was 2–4 mm.

The adaptation of the plates for the bottom appears wholly similar to what it would be in the case of copper plates. Here, too, the metal is worked cold with the wooden mallet. It preserves

the shape given to it without deformation; for pieces with sharp curves and bulgings it is best to take an aluminum that is but slightly alloyed. Aluminum is soft, like copper, and like copper may be bored without difficulty; it is, however, desirable to use tools as sharp as possible, and to oil them before use with petroleum or turpentine-oil.

Nor is there any difficulty in the riveting; the plates may receive a hard hammering without being split, do not turn, stay straight, and do not hollow out at the rivet-holes, so that the rivets hold well even in millings. At most, since it is very malleable, the metal occasionally shows a tendency to bulge out a trifle where the rivets come too near the edge. When the rivets have once been driven in it is difficult to remove them, even with the pliers; they are generally considered weaker than iron, but are placed nearer together.

Aluminum may be filed and grooved like red copper, to which in fact it is similar, indeed, in many respects, except that in case it is desired to harden the metal one must take the precaution to work it cold in so far as possible, after repeated annealing. Alloyed and hammered, it may perfectly well be turned and planed if the instruments are sufficiently sharp and work at sufficient speed. The latter are lubricated with turpentine-oil or petroleum, or, better still, with suds—in no event, however, with oil. The work of milling proceeds smoothly. When the cutters become clogged, as

frequently happens, they must be cleansed with oil and a brush.

Aluminum takes a high polish, but the lustre is not white, as in the case of silver or nickel, but bluish, as with tin. Certain alloys in particular show these hues very clearly. The pieces are first scoured with pumice-stone, and then polished with brushes which have a paste rubbed upon them. The latter consists of half-powdered emery, which is mixed with tallow and formed into small pellets. The polishing is finally completed with polishing-soap and turpentine-oil.

The pure metal, annealed, bends very readily, is easily chased, but does not harden so well, and possesses when it has been worked but little stiffness; the alloys, on the contrary, particularly with 6% of copper, have an unusually great resistance even when chased, but are harder to work. If, however, the material permits of being heated to 100°–150° C., the treatment is thereby made considerably easier. The treatment when cold should be made as brief as possible, in order not to keep the alloy too long under .at a tension.

Processes for Soldering Aluminum.

A great many processes have been devised for soldering aluminum with itself or with another metal; it would appear, however, that up to the present time no truly practical, simple, and well-tested method has been devised. One may set

about the operation in two different ways: either by uniting the surfaces to be soldered by means of a special solder,—hence by means of an easily melted alloy,—or again by a process of so-called autogenous soldering, in which the addition of any foreign metal or of an alloy is avoided. As a rule, the first process is employed.

First Process.—Of the large number of receipts and prescriptions coming under this heading we shall here cite only the most important.

1. Dr. Edward D. Self writes in the "Moniteur scientifique" (1887) as follows: "The great difficulty in uniting two pieces of aluminum is due to the fact that at the place of soldering an extremely thin film of alumina is formed, which resists the union of the metal with the solder in question." With the exercise of great care, however, according to the statement of Self, good results should be obtained with the following alloy:

Type 1: 1 part silver, 2 parts aluminum.

Type 2: 85–95 parts tin, 15–5 parts bismuth.

Type 3: 99 parts tin, 1 part bismuth; with the addition of 1 part aluminum the strength of this solder is much increased.

Type 4: 90 parts tin, 5 parts bismuth, 5 parts aluminum.

The two pieces, well cleansed, are first carefully warmed a little, and the solder is then put on by means of a soldering-iron, vaseline or paraffine serving as a flux.

2. Mourey (1859) gives the following receipts:

Type 5: 80 parts zinc, 20 parts aluminum.
" 6: 85 " " 15 " "
" 7: 88 " " 12 " "
" 8: 92 " " 8 " "
" 9: 94 " " 6 " "

For the production of this solder, first aluminum is melted, and then zinc is added bit by bit with constant stirring. The soldering itself is done with a soldering-iron; the soldering-surfaces are moistened with a composition of 3 parts Copaiba balsam, 1 part Venetian turpentine-oil, and a few drops of a weak mineral or plant acid (phosphoric acid, uric acid), and the zinc, during the whole operation, is protected as far as possible from oxidation.

3. Bourbouze (1866) solders aluminum by tinning the surfaces which are to be joined. For this purpose, instead of employing tin alone, he makes use of various alloys of tin with zinc, with bismuth and aluminum, and with aluminum alone; the latter alloy he considers the best; the quantity of both constituents alters according to the method of the further treatment of the material in question.

The solder

Type 10: 10 parts aluminum, 44 parts tin

is especially recommended. It is sufficiently malleable to permit of being worked with the hammer; pieces soldered in this way may be planed

and turned. If, however, the objects are not to be exposed to any further treatment, a soft solder which contains less aluminum is suitable.

For the tinning itself, Bourbouze indicates no special precautions.

4. A solder with the following composition is said to give very satisfactory results:

Type 11: 5 parts zinc, 2 parts tin, 1 part lead.

5. In the section that treats of light alloys we have given some receipts for the production of soldering-metals which we shall here repeat.

For nickel-tin-aluminum alloys are used

Type 12: 4 parts silver, 8 parts zinc, 5 parts tin (soft solder), and

Type 13: 5 parts silver, 8 parts zinc, 5 parts tin (hard solder).

For partinium:

Type 14: 60 parts zinc, 30 parts tin, 4 parts nickel, 4 parts copper melted with 2 parts potassium arsenate.

6. Charpentier-Page has commercially introduced two kinds of soldering-metal:

Type 15: 48 parts tin, 27 parts zinc, 23 parts lead, 2.25 parts aluminum;

Type 16: 40 parts tin, 100 parts zinc, 20 parts lead;

and he gives the following instructions for their use: The parts are steamed with potash and polished, in order to keep the surfaces perfectly smooth and free from grease; the soldering-iron

is cleansed with the file and smeared with sal-ammoniac; if it is tinned, merely the filing away is sufficient. The surfaces to be soldered must not be moistened with nitric acid nor with any other reagent; they are first tinned with one of the above-named alloys applied to each other, and then soldered as usual with the soldering-iron. When the surfaces have once been tinned with the Charpentier-Page metal, the usual tin-solder also holds very well. Charpentier-Page has by his process even soldered tubes of considerable length.

7. Novel (Geneva) produces soldering-metals with strong resistance, whose composition and method of production are, however, kept secret by the inventor. It is worthy of note that in three out of four tests for strength held at the "Conservatoire des arts et métiers" there was no tearing away at the place of soldering.

8. Wagner recommends the following composition:

Type 17: 100 parts tin, 165 parts lead, 9 parts zinc.

9. According to Lejcal the following alloy gives good results:

Type 18: 2 parts tin, 5 parts zinc, 1 part lead.

10. J. W. Richards, in the "Journal of the Franklin Institute" (1896), publishes the following data:

Type 19: 1 part aluminum, 1 part phosphorus-tin (10%), 8 parts zinc, 32 parts lime.

Type 20: 2.38 parts aluminum, 78.34 parts tin, 19.04 parts zinc, 0.24 parts phosphorus.

Type 21: 2.38 parts aluminum, 71.12 parts tin, 26.19 parts zinc, 0.24 parts phosphorus.

Richards has observed that at the moment of transition into the molten state with these alloys an easily melted compound having the composition 4 parts tin and 3 parts zinc is separated off, which seems to be more durable and solders better.

11. P. d'Arlatan, in the "Chronique Industrielle" of December 15, 1900, has published a number of patented receipts for the production of soldering-metals: the first is the one proposed by S. Tailor in Birmingham.

Type 22: 4 parts aluminum, 12 parts silver, 4 parts copper, 8 parts zinc, 12 parts lead or cadmium, 60 parts tin.

Silver is melted in a graphite crucible, and thereupon the other metals are added successively, in the order given, while the whole is kept constantly stirred by means of a steel rod.

Type 23: 30 parts tin, 50 parts cadmium.

This solder, which is of the consistency of dough, is rubbed upon the previously warmed object of aluminum by means of a piece of asbestos; the soldering then proceeds. In quite the same way are handled also alloys of tin-zinc and of tin-zinc-cadmium (English patent No. 8406). If a soldering-pipe is at hand, common soldering-metal may

also be used, with chloride of silver for the flux. Chloride of silver alone may also be used, if it is applied powdered to the surfaces to be soldered after they have been well steamed; the soldering then proceeds as usual.

It is further proposed, in the publication cited, first to tin the aluminum with an alloy of the composition:

Type 24: 1 part aluminum, 5 parts tin,

and then to solder with the same mixture.

12. Vevey's "Science Pratique" recommends:

Type 25: 45 parts aluminum, 70 parts zinc, 15 parts copper.

13. L. G. Delamothe, chemist, in New York, produces a metal, according to "Nature," which contains

Type 26: 160 parts tin, 40 parts zinc, 10 parts britannia, 10 parts silver.

By "britannia" is understood an alloy containing 100 parts tin, 8 parts antimony, and 2 parts copper. Directly before the cast the crucible is removed from the fire, and 1 g of phosphorus is added, while a stirring is kept up, by means of an iron rod, till the phosphorus is completely consumed. The alloy is then cast in bars, with which the parts to be soldered are tinned; the soldering itself may be undertaken either with the same alloy or with common solder, with the aid of a solder-pipe or soldering-iron. The surfaces must previously be brushed off, and

thoroughly cleansed with rosin dissolved in stearin.

14. Professor C. D. Thiving of Knox College recommends an alloy of zinc, tin, and bismuth, which melts at a very low temperature and may therefore be conveniently handled with the soldering-iron. It is affected neither by water nor by damp air, and as regards mechanical resistance is scarcely inferior to aluminum.

15. Gaston Tissandier, in his "Recettes and Procédés utiles," refers to the fact that if both metals are previously coppered galvanically, or by some other means, the soldering may easily be effected, even by the ordinary means, except that the greatest care must be taken to see that the copper coating melts or separates off. As regards the coppering, Tissandier remarks that in cases where it is impracticable to immerse the parts to be soldered directly in a copper bath, very good results may be obtained by the use of blotting-paper saturated with copper sulphate. As soon as the contact of the paper with the surface in question and with a piece of copper has been brought about, the positive pole of a battery is connected with the latter, while the negative pole is connected with the aluminum object, and after a short time a firm coating of copper is obtained on the surface to be soldered. A mixture of rosin, tallow, neutral zinc chloride, and quicksilver sublimate is also said to serve the purpose well. A suit-

able solder will be found in an alloy of the composition:

Type 27: 52 parts copper, 46 parts zinc, 2 parts tin, with borax for the flux.

16. In his "Recueil des procédés modernes" Marcel Bourdais gives more than twelve different methods of soldering for aluminum bronze and aluminum, of which we shall here cite the most important. According to his directions also, the metals must first be tinned with tin-aluminum.

	Aluminum.	Copper.	Zinc.	Tin.	Lead.	Nickel.
Type 28....	30	20	50	....	....	....
" 29....	....	....	....	95	5	....
" 30....	....	....	....	97.5	....	25
" 31....	9	6	85	....	....	....
" 32....	20	15	65	....	....	....
" 33....	....	....	5	95	....	....
" 34....	12	8	80	....	....	....
" 35....	6	4	90	....	....	....
" 36....	75	....	....	25	....	....

17. To Delécluse we owe a very interesting investigation of the processes for soldering aluminum and the methods of preserving the elasticity of the metal throughout its various manipulations.

18. Ludovic Olivers, by means of a heated cast-iron plate, heats the aluminum to a temperature of 230°–250° C. He then rubs in hard upon the parts concerned a solder whose composition he does not state, taking care to see that the solder is evenly distributed, by means of a metal brush. The solder

melts, and is properly deposited on the surfaces. In order to remove the last traces of oxide, the soldering-places are finally rubbed off once more with care, the two surfaces are put together, and the soldering finished in the usual manner with the soldering-iron.

Otto Nicolai uses a zinc-tin solder, or some other alloy generally employed for this purpose, and secures a permanent union by covering the soldering-places with powdered cadmium chloride or cadmium iodide.

20. In a very exhaustive paper of W. S. Bates before the Chicago meeting of the American Chemical Society on March 18, 1898, he recommends the alloy:

Type 37: 70 parts aluminum, 30 parts tin,

which would assure a very secure union, were it not for the fact that in time the alloy is subject to molecular action which impairs its strength.

The alloy

Type 38: 81 parts aluminum, 19 parts copper

melts sooner than the preceding, it is true; but it is not subject to any molecular alteration, and likewise makes a firm solder.

Bates has obtained his best results with a ternary alloy of the composition:

Type 39: 70 parts aluminum, 20 parts tin, 10 parts copper or silver.

The mass-proportions of these metals may vary between wide limits, but the composition cited is

the best. These alloys solder excellently and very firmly. Some of them in the course of a year showed no such alteration as is usually observed; others, which had been experimentally placed in water for a month, showed themselves fully capable of resistance, revealing no trace of a galvanic effect.

For a flux, in the case of soft solder, which may be worked at a relatively low temperature, Bates uses zinc chloride, stearin, soap, sugar, quicksilver chloride, and certain iodides; for hard solder, which melts only at a higher temperature, borax, alkali fluorides, lithium chloride.

21. Grant Hammond in San Francisco proposes for a flux a mixture of iodine, tin iodide and quicksilver iodide, and a hydrocarbon from the vaseline series. F. A. Gooch proposes a mixture of sodium fluoride, aluminum fluoride, and aluminum-sodium chloride; also a composition of aluminum fluoride, sodium fluoride, and zinc chloride.

Grant Hammond, finally, prefers the solder:

Type 40: 100 parts tin, 20 parts silver, 10 parts zinc, 1 to 6 parts aluminum,

to all similar compositions.

Second Process: Autogenous Soldering. — In the case of aluminum, great difficulties are met with in attempting to solder without the aid of a foreign metal or of an alloy.

1. Spring has shown that two carefully worked pieces of aluminum, if pressed together and heated to 420° C., in the course of about eight hours

show signs of fusion. If one of the pieces be placed in a vise, the other may be worked without breaking off.

2. Bourgoin has devised a number of soldering processes which partly belong in the group of tin-soldering processes, and partly may be classified in the series of autogenous processes. We mention these for the sake of completeness.

3. We owe to the firm of Heraeus in Hanau a. M. the following method. The surfaces of the two pieces of metal which it is desired to unite are thoroughly cleansed over an area of 5–10 mm, and are placed in contact in this perfectly bare condition; the metal is now heated by means of the soldering-pipe until it is of a perfectly fixed consistency. When the required temperature is reached, it is thenceforward kept constant, and meanwhile the two pieces pressed together are hammered by a peculiar method until they are completely fused together. After the cooling down, the union is so firm that it is loosened neither by being suddenly shaken nor by changes of temperature.

Electroplating of Aluminum.

In spite of the great number of experiments in the electroplating of aluminum, we have not, up to the present day, succeeded in obtaining a certain method of coppering, nickeling, gilding, and silvering the metal.

Coppering. — In an exhaustive paper Margot refers to the fact that the most essential device for securing a permanent deposit of copper on aluminum consists in freeing the metal from the coating of oxide with which, of itself, it is already covered, and which continues to form even in the copper bath. With this end in view, the following process is suggested:

1. The cleansing of the aluminum with alkali carbonate, in order to keep the surface rough and porous.

2. The metal is thoroughly washed in running water, and then immersed in a warm solution of about 5% hydrochloric acid; it is thereupon cleansed with pure water.

3. The plates are kept immersed in a moderately concentrate, slightly acidified copper-vitriol solution until an even coating has formed.

4. They are thereupon thoroughly washed again, until the last traces of chlorine have been removed, and finally

5. They are placed in the galvanic bath, where the coppering takes place by means of the electric current.

The "Société électrometallurgique française" in coppering aluminum avails itself successfully of a process which is very like that just described, the only difference being that, in the coppering by mere immersion, the copper sulphate is replaced by a slightly acid copper-chloride solution.

Gilding and Silvering.—It may be said that since

the first experiments in this direction there is no notable progress to be recorded. The process of Lejcal gives very good results, provided only thin metallic coatings are required. The objects to be silvered are heated to about 300° C. and rubbed with a bit of wadding covered with tin chloride; in this way we obtain a permanent coating of tin, upon which the silver is evenly deposited.

The gold and silver baths of the *Société Vienne frères* are likewise said to have acted very well. "La lumière électrique" * contains the following information regarding them: The pieces which it is desired to solder are first steamed with nitro-hydrochloric acid or nitric acid, and then warmed to 70°–80°. The place of the nitric acid may be taken by other similarly acting reagents; nitric acid is, however, the most suitable and practical. Finally, after the pieces have been cleansed with fine pumice-stone in the usual way, they are immersed in baths of the following composition, warmed to about 20°–35° C.

Silver bath: 30 parts silver, 60 parts cyanide of potassium, 1000 parts distilled water.

Gold bath: 7 parts gold, 49 parts sulphurous carbonate of soda, 23 parts cyanide of potassium, 23 parts sodium phosphate, 1000 parts distilled water.

Margot and Minet were the first to suggest that the aluminum be previously covered with a thin,

*"La lumière électrique," July 11, 1891.

firm coating of copper, according to Margot's method mentioned above, and that the metal be silvered or gilded only after it has been thus prepared; in this way aluminum may be nickeled as well, and, in fact, may be plated with any metal that is capable of being permanently deposited on copper.

But with the exercise of care it should be possible, in my opinion, to silver aluminum directly, by the following method.

That the direct silvering of aluminum has hitherto scarcely been successful may be due to the fact that the process was carried out with neutral or alkaline baths. Under these conditions the metal has a tendency to cover itself with a film of oxide, which is not dissolved again, and which hinders the attachment of the coating of silver. If, on the other hand, the parts concerned are previously prepared and steamed, as Margot suggests for the coppering, and if for the bath-fluid a solution of sodium-silver double fluoride slightly acidified with hydrofluoric acid is used, very good results would certainly be obtained.

In view of these considerations, the following method recommends itself. In order to keep the surfaces of the aluminum objects rough and porous, they are first of all cleansed with a diluted, warm solution of alkali carbonate, and then thoroughly washed, immersed in a warm solution of hydrofluoric acid, and only then suspended in the silver-

fluoride bath; any alumina that may be formed is immediately dissolved by the hydrofluoric acid. Under these conditions, when the current is passed through, a very smooth and uniform silver coating is obtained. If the object in question has such dimensions as to be steamed with difficulty, it is sufficient to connect it as anode in the silver bath for a few seconds, while the fluorine separating off provides for the steaming. After a short time the direction of the amount is changed, whereupon the silvering begins.

A general method for obtaining **metal platings of various sorts,** according to Golting, consists in immersing the aluminum in the metal-salt solution in question, and at the same time keeping it in contact with a chosen metal of such a kind that the resulting combination forms a voltaic pile in which the aluminum is the negative pole.

Wagner has mentioned another process: Aluminum is subjected to corrosion in a bath which contains acid copper acetate, iron oxide, sulphur and aluminum chloride; it is then rubbed off with a brass brush, by means of which a metal covering is formed, which removes the coating of grease, stops up the pores, and levels the surface. Next it is thoroughly washed with pure water, and finally immersed in the electrolytic cell. The method is said to give coatings capable of great resistance and durable.

According to Lenseigne and Leblanc, the alu-

minum objects to be plated are steamed in dilute sodium lye or dilute potassium lye or in hydrochloric acid (1:10), then thoroughly brushed off in water, and finally heated, according to the sort of metal coating desired, in one of the following baths:

Gold bath: 40 parts gold chloride, 40 parts cyanide of potassium, 40 parts sodium phosphate, 2000 parts distilled water.

Silver bath: 20 parts silver nitrate, 40 parts cyanide of potassium, 40 parts sodium phosphate, 1000 parts distilled water.

Copper bath: 300 parts cyanide of copper, 450 parts cyanide of potassium, 450 parts sodium phosphate, 8000 parts distilled water.

Nickel bath: 70 parts nickel chloride, 70 parts sodium phosphate, 1000 parts distilled water.

The temperature of these baths should be 60°–70° C., and should be kept constant throughout the operation; for the anode the metal may suitably be chosen whose salt is dissolved in the electrolyte.

D. Uses of Aluminum.

Not merely as an alloy, but in the pure state as well, aluminum has been employed in a great many different ways in commerce and in petty industries, in large enterprises, in chemistry and in metallurgy.

(*a*) Aluminum in Commerce and Minor Industry.

Here belong a large number of articles, useful or luxurious, such as keys, visiting-cards, thimbles, brushes, combs, letter-cases, cigar- and cigarette-cases, etc., for which aluminum is highly valued on account of its lightness; aluminum also serves for the manufacture of opera-glasses, spectacles, knives, watches, articles of adornment; likewise for cooking and household utensils, cups, dishes, chafing-dishes, tea-urns; for military equipment, etc. For articles of small or moderate dimensions, where no special strength is required, this metal already competes successfully with copper, nickel, German silver, and brass, since volume for volume it is cheaper. But also

(*b*) In Greater Industry

aluminum has found a wide field of usefulness. First in importance, in this connection, is cast aluminum, as to the weight and the use of which the last Paris Exposition (1900) gave one a very good idea. Maxime Corbin, the discoverer of an aluminium alloy with a strong resistance, had produced a non-separable mountain gun-carriage weighing only 78 kg, while that adopted by the French army weighs 285 kg; the same exhibitor showed a model for a bed-plate for an 80-horse-power dynamo machine; the weight of this plate amounted to 185 kg, while a similar construction in steel would

weigh 380 kg. Moreover, Corbin has already supplied 150 pieces of machine-parts of this sort for electric vehicles.

Partin with his partinium obtained cast aluminum parts which had an elasticity of 18–20 kg per mm^2. Single machine-parts of the sort at the Exposition aroused a lively interest in professional circles, especially a stand for a 50-horse-power steam-engine, with a weight of only 75 kg. The casting of this large piece containing over a cubic metre succeeded perfectly; there were no blow-holes or other defects, such as often occur in castings.

The automobile and the bicycle have, of course, opened up a new field of usefulness for the alloys of aluminum, and particularly partinium; of this, too, the last Exposition of 1900 afforded striking proof in the exhibition of numerous automobile parts, as well as speed-regulators, casters, friction-wheels, and large and small models of every sort.

The visitor's attention was particularly arrested by an aluminum bell, 70 cm in height, 50 cm in greatest width, and weighing about 15 kg; on being struck it gave forth a sound which did not differ from that of a bronze bell. Of the other objects exhibited by Partin we may mention: an anchor used on several balloon-trips by Messrs. Hervieu, Mallet, Gilbert, and Variclé, which was found particularly serviceable by Hervieu in one of his perilous voyages in Russia; a pillar with capital, which afforded a demonstration of the use

of aluminum for ornamental purposes; a chased cup, deceptively similar to old silver; a statuette, an ash-tray, a Psyche, a beautifully worked hand-mirror, a wash-basin, a water-pitcher, etc.

We have already emphasized the use of aluminum as material for conducting-wire; with equal conductivity it weighs and costs less than copper. Aluminum also permits of being worked up into tubes; such tubes are preferred for optical instruments. An attempt has also been made to introduce aluminum tubes in the manufacture of bicycles; the frames were to be made of a light alloy to which it was designed to give the greatest possible strength by means of careful casting, remelting, and any further treatment required; I believe, however, that aluminum will be found to associate itself with steel as reluctantly as do other metals, in those cases where lightness and an extraordinary power of resistance are desired.

Rolled aluminum finds its chief application in ship-building and for military purposes. The first to produce rolled aluminum plates of large dimensions for ship-construction was Charpentier-Page, who also was the first to point out the enormous advantage of aluminum for field-equipment utensils, and who together with Japy devised many and various types of these articles.

The countless mechanical tests which were made by Charpentier-Page on aluminum and copper-aluminum alloys we have already mentioned. We

may add here the results of some tests which concern themselves with an alloy having a high power of resistance, and it is an alloy that, from a technical point of view, is most useful.

HARD WIRE.

2 mm in diam. Density 2.98.

Electrical Tests.

Resistance per metre	0.01598Ω
Resistance per km length and mm² cross-section	50.17Ω
Copper wire of the same dimensions, at 15° C	17.4Ω
Proportion of conductivities	34.6%

Mechanical Tests.

	Test.		
	1	2	3
Length of test-piece.....mm	105	105	105
Elasticity..... kg	140	139	137.65
Elasticity per mm² cross-section..... kg	44.57	44.2	43.82
Elongation.....mm	3.5	3.5	2.9
Elongation..... %	3.3	3.3	2.7

The usefulness of aluminum for military purposes was demonstrated at the Exposition by a most interesting object, namely, a movable bridge of aluminum, which the Sedan works had exhibited in the "Palais des Armées de Terre et de Mer." It is a portable bridge which was constructed under the direction of General Dumont and according to the plans of Commander Houdaille. Its span is 15 m; it consists of three lengthwise beams which together weigh 900 kg; the weight of the bridge-load is 600 kg, so that the entire bridge weighs

1500 kg. Its maximum load is officially given as 9000 kg, i.e. 600 kg per metre of length. With this load the sag is 70 mm, with the bridge unloaded it is 22 mm.

The bridge is strong enough to bear a wagon with six horses, weighing together 2300 kg, and forty men.

Field-equipment Utensils. — After the autumn manœuvres of 1894 an explicit report was made to the French minister of war regarding the experimental introduction of certain aluminum utensils as articles of equipment. The articles comprising the aluminum cooking apparatus, of three different sizes, weigh 540, 285, and 50 g respectively. The so-called small equipment, in which the same objects weigh 385, 215, and 40 g respectively, it is true, gave less satisfactory results in the tests, for while the large equipment completely satisfied all requirements during the manœuvres, the small equipment soon became unserviceable.

Since the collective weight of the iron cooking apparatus, such as is still used in the French army, is 1385 g, as compared with 875 g for the weight of the aluminum utensils, in the latter case the soldier's burden is lightened by no less than 510 g. Other countries besides France—Germany, Russia, and Austria among them—have made thorough tests, and have in part adopted aluminum utensils.

Ship-construction.—The first vessel in which aluminum was used in considerable quantities was the "Vendenesse" of Count J. de Chabannes La Palice,

which, built after the plans of Victor Guilloux of Godinot, sailed from St. Denis in December, 1893. Throughout its voyages the behavior of the vessel was carefully noted by Guilloux, so that we have valuable observations as a result, of which we shall proceed to give the more important.

Three months after sailing the vessel was subjected to a thorough inspection at Havre, and was found to be quite intact in its interior. On the outside, in a few places where the paint had been damaged in sliding down the ways or during the passage down the Seine, the rivet-heads had become slightly oxidized; and the bare spots showed, in addition to a fairly uniform but otherwise insignificant oxidation due to the formation of alumina, a few more serious injuries. The vessel was repaired and painted, and soon after continued its journey. When about two months had elapsed it received another overhauling at the dock at Honfleur. Guilloux found the vessel on this occasion in perfect condition; even in those places where mussels had become attached to the hull and the paint was gone, the aluminum had retained its lustre unimpaired. Only upon the deck were noticeable here and there the signs of incipient oxidation; the deck, to be sure, was not painted, but merely covered with oilcloth, most of which had worked loose.

Even upon a third professional inspection the interior of the "Vendenesse" was found to be

wholly undamaged; except that in the places where the water had worked its way in beneath the wooden planking there was a clearly perceptible yet unimportant effect upon the metal. The deck, on the other hand, throughout its entire extent was covered with a layer of alumina, fragments of oilcloth and iron-lime, which, carefully collected, dried, and analyzed, gave a total weight of 8 kg, including 5 kg of aluminum. This, calculated for the entire surface of the deck (20 sq. m), expresses a loss of 0.1 mm, which could have been avoided by painting, and which, moreover, would not result in any serious inconvenience if the oxidation were but equal in amount over the entire surface, so that the durability of the metal parts concerned might be previously determined. Unfortunately, this is not the case, for while some of the riveted plates of which the deck consisted were equally affected, others showed very irregular unevennesses and depressions; the plate farthest aft was most affected; the parts in the neighborhood of a (copper) ventilating device had also suffered severely.

The parts which were most oxidized were filed till the coating of alumina was entirely removed, and were then immersed in pairs in a solution of sodium chloride, in order to measure the electromotive force of the chain eventually formed. It amounted to 0.05–0.10 volts, and attained its highest value when the most oxidized plate was taken as the cathode. It follows from this that in making

the deck plates not chemically identical were used, so that it was possible for local currents to be formed which resulted in oxidation. In ship-construction, therefore, it is a most essential condition that only such aluminum shall be used as has become wholly homogeneous by means of a series of careful re-meltings.

How important a condition this is may be seen from the fact that the torpedo-boat "La Foudre," which the French government had built by the English firm of Yarrow shortly after the trial trips of the "Vendenesse," quickly went to pieces, since sufficient care was not taken that homogeneous metal should be employed exclusively.

Among other aluminum vessels we may mention:

The portable boats of Lefebvre (built jointly with Guilloux) which during the last few years have been despatched to African waters, and have there done good service. A model was exhibited at the World's Fair of 1900: the portable "Etienne" (10 tons), with a total weight of 1050 kg, built in the year 1893, restored by Colonel Marchand after a three years' voyage on the Congo. On this vessel Commander Baratier explored the whole of Bakr-el-Ghazal, and Marchand made the journey to Fachoda (July 10, 1898). This expedition was accompanied by the two vessels "Commandant Besançon" (8 tons, 400 kg in weight) and "Jules Davoust," both of which were built in the year 1893.

The vessels "Crampel," "Lauzière," and "Plei-

gneur" (13 tons) were still in use in 1900 on the waters of the Congo, while "Grall," "Livrell," "Pronci," and "Jansaric" (50 tons; built 1894) were plying to and fro upon the Niger.

The materials for these vessels, aluminum plates, were furnished chiefly by the factories of Charpentier-Page and the Sedan Works, under the direction of Dreyfus, Paris representative of the "Société electrométallurgique française." Several pleasure-yachts were also built in Germany and in Switzerland. Escher Wyss & Co. in Zürich exhibited at the World's Fair a vessel with a two-horse-power petroleum motor, which weighed 400 kg and was able to cover 5–6 knots per hour; also a small boat for four persons, weighing but 48 kg.

Of racing-boats we may mention: the "Luna" of Mr. Arons (5 tons) and the "Alumin" of Mr. Huldschinski (10 tons), both in Berlin. The racing for the "America" Cup in the year 1895 is still fresh in the minds of all, in which the American yacht "Defender" and the English "Vigilant" took part. The latter was sheathed with plates of Tobin bronze, the former with plates of bronze-aluminum.

That aluminum has rendered good service in aeronautics as well, we have already remarked: in this connection it serves principally for the manufacture of the balloon-car; and it has been employed for this purpose most advantageously, since this inner stiffening has made the balloon much

better able to withstand storms and accidents, and much more reliable. The Russian engineer Tschernouchouko has built a balloon of this sort, which with a weight of 4800 kg was able to lift 100 men and a ton of baggage.

(*c*) Aluminum in Chemistry and Metallurgy.

Aluminum is much used as a reducing-agent in melting cast iron, in refining steel, in the production of certain metals, in aluminothermy, so called; also in the production of phosphorus and in photochemistry.

The Production of Phosphorus. — Accidentally, during his investigations as to the possibility of soldering aluminum, Professor Rossel in Bern observed that phosphates are reduced by aluminum. When Rossel heated a mixture of aluminum-foil and phosphoric substances (phosphates) in a porcelain crucible, he observed that little flames spurted out at the side of the melt. He repeated the experiment in a closed tube, in order to avoid the oxidation of the vapors passing off, and could thus easily demonstrate the presence of phosphorus in the product of reaction, with a consumption of about 30% of the original material. In order to reduce all the phosphoric acid to phosphorus, to the mixture of aluminum and phosphates (or, better, metaphosphates) silica had to be added.

In **Photochemistry** aluminum may be employed in two ways. According to the suggestion of the French chemist Clemmon, gold and silver from photographic baths which can no longer be used are to be precipitated in such a manner that an aluminum plate may be immersed in the solution, the latter having been strongly acidified with hydrochloric acid; gold is separated off directly, with the formation of abundant gases; silver is precipitated as a chloride.

A second theory originates with Professor Glusmaff. His idea is to use aluminum instead of magnesium as a source of light for taking photographs in the dark; and he gives the following receipts:

1. 21.7 parts powdered aluminum, 13.8 parts sulphide of antimony, 64.5 parts potassium chlorate.

The combustion of this mixture lasts but $\frac{1}{17}$ second.

2. 30 parts powdered aluminum, 70 parts potassium chlorate.

This mixture dies out within $\frac{1}{5}$ second.

Aluminum has still another chemical function, as *reducing-agent in refining cast iron, steel, and other metals*. The formation of an oxide, which happens during the melting of many metals in air, is prevented when aluminum is added, on account of the reductive properties of the latter; certain impurities present in the metals are likewise reduced, so that a casting free from blow-holes is obtained, which in

consequence of the elimination of the oxides likely to be enclosed is neither fissured nor brittle. According to Langley, aluminum is to be added in the following quantities, according to the nature of the metal to be refined:

0.016–0.030% Al to Martin steel with 0.5% C.

0.020–0.050% Al to Bessemer steel with 0.5% C.

0.011–0.025% Al to Bessemer steel with more than 0.5% C.

The Aluminium-Industrie-Aktien-Gesellschaft refines

steel	with	0.004–0.025%	aluminum
soft steel	"	0.01 –0.1%	"
cast iron	"	0.2%	"
copper	"	0.1 –0.25%	"
brass	"	0.1 –0.50%	"
nickel	"	0.027–0.09%	"

Foucau is of the opinion that in the metallurgy of iron it will be possible to avoid entirely the production of carboniferous ferro-silicon, and to limit to a considerable extent the use of ferro-manganese, since aluminum is destined to replace the former altogether, and the latter in those cases which are not directly concerned with the separation from sulphur. In the case of nickel-steel, aluminum may likewise be employed to advantage, since it considerably simplifies the casting. With other metals as well as iron, very pure and uniform castings are obtained with the aid of aluminum, castings

which satisfactorily withstand the effect of cold or of heat.

Keep, engineer of the American The Michigan Stove Works Company, jointly with Mabery and Vorce has investigated very thoroughly the effect of aluminum upon molten iron, and has been able to demonstrate that aluminum changes into graphite the carbon that is in chemical union and that which is dissolved, and in some peculiar way hinders the carbon from raising blisters; on the contrary, it brings about the even distribution of the carbon throughout the mass at the moment of cooling.

The effect of various quantities of aluminum upon white cast iron in particular is given in the following table:

Adding	0%	aluminum,	white fracture.
"	0.25%	"	grayish-white fracture.
"	0.50%	"	lustrous gray fracture.
"	0.75%	"	gray fracture.
"	1.00%	"	dark gray fracture.

0.25% aluminum, according to Keep, is equivalent to 0.62% silicon, in so far as the nature of the surfaces of fracture is concerned.

The effect of aluminum on iron carbide has been carefully investigated by T. W. Hogg. He first points out that in these carbides so many foreign elements are contained, in such various proportional quantities, that it is exceedingly difficult to determine precisely the rôle of each in the dif-

ferent modifications of the carboniferous iron exposed to the effect of the aluminum. The difficulty of such an investigation is all the greater if we take into account the effect which certain special conditions, such as the variations in the melting-point and the rapidity with which the cast iron stiffens, have been found to produce.

Another circumstance which materially affects the readiness with which the carbon passes from the chemically united into the graphitic state is the amount of carbon, in particular, contained in the iron. In this respect the iron may be compared to some extent with a more or less saturated salt solution. T. W. Hogg has made numerous experiments in the effort to throw some light upon this question, which has hitherto been but little investigated. He found that the addition of 1% of aluminum considerably modified the carbon which was in chemical union, since it changed the latter into graphite. With the addition of more aluminum the carbon gives evidence of a tendency to return again to its original condition; in the presence of 12% of aluminum it has usually returned to that state.

In a paper appearing in September, 1891, which treats of the use of aluminum in refining steel, Le Verrier, Professor at the "Conservatoire des Arts et des Métiers," comes to the following conclusions:

1. Aluminum is an energetic reducing-agent, and

although it is difficult to oxidize it directly, it reduces almost all metallic oxides in the heat.

2. Aluminum causes soft steel in the molten state to flow easily.

3. Aluminum prevents blistering better than any other addition.

Aluminum as Reducing-agent in the Production of Metals and Alloys.—We have already (page 171) cited an example for the production of an alloy (chromium-aluminum) by the reduction of the salt concerned with aluminum, according to Wöhler, who in fact was the first to use metals as reducing-agents in pyro-chemistry.

Charles Combes has published a very interesting paper dealing with this subject, which he laid before the "Académie des Sciences" on June 25, 1896. He succeeded in producing the following alloys:

Nickel-aluminum, with 20% Nickel.—Aluminum is allowed to work upon nickel sulphite (NiS); the reaction takes place with violent boiling.

Manganese-aluminum. — Anhydrous manganese chloride is introduced into molten aluminum; there are formed aluminum chloride, which quickly evaporates, and may be received in a condenser, and an alloy containing 4% manganese, with the fracture crystalline and similar to specular cast iron.

Chromium-aluminum.—Aluminum and chromium chloride (with a violently foaming reaction) give the alloy already discovered by Wöhler. With 7% of chromium it is brittle and has a crystalline

structure; with 13% of chromium it is perfectly crystalline, and may be powdered in mortars.

Aluminothermy.

Up to this point we have only described such processes for the production of aluminum alloys as depend on the reduction by means of haloids or sulphides. Besides these there is a further group of methods which may be classified under the term "Aluminothermy," and which have for their object the reduction of metallic oxides.

As early as the year 1885 the brothers Tissier attempted to reduced manganese by means of aluminum, without, however, arriving at any result. Later they heated an equivalent of iron oxide with three equivalents of aluminum to a white glow, and thus obtained amid explosive phenomena a metallic regulus with 60.3% iron and 39.7% aluminum.

Copper oxide also is reduced by aluminum at a white glow; in the case of litharge the reduction takes place at precisely its own melting-point, and so violently that the melt may be tossed up out of the crucible so that it raises the roof of a small Perrot gas-furnace. The same phenomenon was also observed by Boussingault, and Richards mentions it in his work on aluminum.

Ritto has reduced uranium oxide by means of granulated aluminum, and thus obtained an alloy with 65% uranium and 35% aluminum. Moissan, among other things, has succeeded in introducing

the metals hardest to fuse, such as molybdenum, tungsten, titanium, chromium, etc., into pig iron, cast iron, steel, and bronze, with the aid of alloys which were produced by means of the direct reduction of the oxide concerned with aluminum or some other flux. The reduction of silica as well was made possible in this way. If two molecules SiO_2 and four molecules Al are thoroughly mixed and carefully heated, at 800° C. the reduction from silica to silicon takes place.—The author has obtained a metal very rich in silicon by introducing potassium silicate into melted aluminum.

Researches of Hans Goldschmidt.—While all the investigators hitherto mentioned obtained only aluminum alloys, Hans Goldschmidt was the first to produce pure metals by means of the reduction of certain oxides; he may, therefore, properly be called the father of aluminothermy. Many experiments before the time of Goldschmidt had, indeed, taught that the reduction of certain metallic salts and metallic oxides takes place with extreme violence, sometimes even explosively; there was lacking, however, any definite information as to the height of temperature of the reaction. It was not yet known that the temperature of combustion of aluminum was sufficiently high to melt alumina and even chromium without the presence of a flux,—the latter metal, as is known, being among the most refractory; up to that time it had been found infusible even in the electric arc.

If one compares the heat of combustion of certain other elements, as given in the tables of Landolt and Börnstein, with that of aluminum, as it was obtained by Dr. Strauss, physicist of the firm of Friedrich Krupp in Essen, we have the following instructive figures, which we take from a reference of Goldschmidt's:

Element.	Heat of Combustion.
Hydrogen	34000 cal.
Carbon	8317 "
Aluminum	7140 "
Magnesium	6077 "
Phosphorus	5964 "
Sodium	3293 "
Calcium	3283 "
Sulphur	2200 "
Zinc	1314 "
Copper	321 "
Silver	27 "

If operations are carried on with large quantities, as was Goldschmidt's intention, a twofold difficulty is met with. In the first place the violence of the reaction must be lessened as much as possible, and in the second place a crucible material must be found which is not affected by aluminum in the molten state. The reaction must take place in such a manner that the alumina as it forms is deposited

upon the inner surface of the reaction-vessel, so that the receptacle is protected in and of itself from further attack through the formation of the layer of alumina.

Another point was to decide the question, theoretically and practically of equal importance, whether it was possible so to conduct the reaction that the combustion once begun should continue of itself without any further addition of heat; and whether, indeed, the combustion might not take place either *cold*, provided that the mix is in and of itself able to maintain the combustion, or *warm*, that is to say, in such a manner that the mix is first brought to a temperature favorable to the further spontaneous course of the reaction, and is then left to itself. This temperature, of course, could only have been determined experimentally.

One may easily imagine the technical difficulties which stood in the way of carrying out this aluminothermic process, —difficulties which, indeed, could never have been overcome if in practice it had proved necessary as a preliminary to heat the mixture to the temperature of combustion by external means.

The first metal which Goldschmidt endeavored to produce aluminothermically was chromium. After he had ascertained that chromium oxide may be reduced by means of aluminum, he mixed both substances thoroughly in a crucible, and attempted, at first with a slender flame, to kindle the mixture

locally; only after repeated unsuccessful attempts, however, did he succeed in discovering the conditions under which the mixture actually burns and the reaction, after the point of combustion has been reached, proceeds in such a manner that it may easily be controlled. A new method of thermochemical investigation was revealed by these pioneer researches, and at the same time the foundations were laid for a new branch of industry—aluminothermy.

It now became merely a matter of developing the method technically, especially with reference to starting the reaction. It followed that for this purpose a mixture of aluminum and such oxides or superoxides as gave off their oxygen more readily than chromium oxide was best adapted. This combustible material, with the purpose in view, was placed within the reaction material in a small hollowed-out space, and was set on fire by means of a slender flame; the reaction began within a minute or two thereafter. In place of the flame a ring of magnesium at once appears, which is brought to combustion simply by a lighted match. The inflammatory mixture which Goldschmidt employs at present, which he calls "Zündpatrone," has the form of a small pellet of powdered aluminum and barium superoxide, to which a strip of magnesium is added. Upon the amount of combustible material added, the ease with which the course of the reaction is regulated depends.

In this way Goldschmidt succeeded in obtaining several kilograms of chromium at a single operation, and, as early as 1894, he obtained at each charge not less than 25 kg of this metal.

Further experiments have shown that a large number of metals, especially manganese and iron, behave like chromium. A part of the aluminum in the operation may be replaced by magnesium or calcium carbide; in the latter event the reduced metals are more or less carboniferous.

Likewise in place of the oxides sulphides or other metallic salts, especially sulphates, may be used. A mixture of pure iron oxide and aluminum gives wrought iron directly.

Technically, Goldschmidt's process may be employed in three different ways:

1. For the production of pure metals.
2. " " " " corundum.
3. " various thermic purposes.

1. The Production of Pure Metals.—Experiments have shown that even with a very slight excess of metallic oxide metals or alloys entirely free from aluminum may be obtained. This fact is very remarkable, on the one hand because aluminum possesses the property of being alloyed with extreme ease, on the other hand because, as we know, reduction by means of carbon never results in metals free from carbon being obtained.

Goldschmidt produces at present in his works at Essen about 100 kg of chromium at a single charge; the melting-crucibles must of course be so constructed that they are able to withstand the pressure of this mass. The work of the operator is confined to introducing the mixture of chromium oxide and aluminum; the production of 100 kg of chromium, it is stated, takes twenty-five minutes; the metal obtained is entirely free from carbon.

The operation in the case of manganese is altogether similar; manganese, produced aluminothermically, comes upon the market at present in large quantities, of an extraordinary purity. Titanium also is produced by the Goldschmidt process, not, however, as pure metal—on account of its high melting-point,—but alloyed with iron; furthermore, barium oxide and lime may be reduced with aluminum.

In order to convince himself of the universal applicability of his process, Goldschmidt has also attempted to reduce most of the other metal oxides with aluminum, without, however, obtaining in all cases a well-defined reaction. It is interesting to note that vanadium acid up to the present time has withstood this treatment, which in this case resulted merely in the lower stages of oxidation, and not in the production of pure metal.

2. Production of Artificial Corundum. — The slag occurring in the melting-crucible simultaneously

with the formation of metal is nothing else than molten alumina or artificial corundum, which, to distinguish it from corundum otherwise originated, is called *corubis*, and whose simultaneous production considerably increases the economical value of the aluminothermic process.

3. The Generation of High Temperatures; Soldering.—While heretofore it was necessary in welding to employ, even for the very smallest flange, a charcoal fire or a water-gas flame, the work of soldering is by Goldschmidt's process greatly simplified. Let us suppose, for example, that it is desired to solder a flange upon an iron tube an inch in diameter. The solder is introduced betwixt the parts by means of borax; the flange is enveloped in a paper covering, which should be somewhat wider than the external diameter of the disc, and on top and at the sides the latter is covered with a layer of sand; whereupon the whole is placed in a receptacle of sheet metal of the proper shape and size. In place of the paper covering thin iron plate may be used. The flange is then immersed in the heating mixture, so that the parts to be soldered are completely covered; the mixture is enkindled, and finally dry sand is poured upon it. The effect of the heat developed is to melt the solder, and thus to bind both the iron parts firmly together. The slag formed during the process is not removed, of course, until it is completely cooled.

The Goldschmidt method is not yet available in all cases, nor have its possibilities been fully developed; but already it is realized that this new process is capable of extremely interesting and manifold applications.

APPENDIX.

SUPPLEMENTARY NOTES BY ADOLPHE MINET.

In the course of the two or three months following the publication of the German edition, several criticisms were made which the author desirous of accuracy and impartiality could not, in justice to himself, pass over in silence.

These criticisms, at least the chief ones, appeared successively in the *Zeitschrift für Elektrochemie*, over the signature of M. Haber; in *Electrochemical Industry;* and in *The Electrochemist and Metallurgist.*

Since the majority of these criticisms have concerned themselves with the same points, and the German criticisms have been the most numerous, this additional chapter has been written mainly with a view to answering the criticisms of the *Zeitschrift für Elektrochemie.*

For the sake of brevity, I will not reproduce the positions of the text under discussion, but I will refer the reader throughout the course of the

controversy to the English text, which is an exact translation of the German.

Some of the criticisms concern themselves specifically with the industrial, others with the theoretical aspects of the question; we shall, therefore, make a twofold division of our subject-matter.

Industrial Questions.

1. Mr. Haber informs the readers of the *Zeitschrift* of his disappointment, in reading the German version, at not finding the valuable information as to the preparation of aluminum he had hoped to find.

The electro-metallurgy of aluminum being a comparatively recent industry, and one which has been put on a solid basis only after considerable experiment and expenditure of time and money, it seemed to lie outside my province to give the specific details of manufacture, as Mr. Haber would have liked, and I deemed it best to restrict myself to questions of a general nature. The right to do otherwise belongs to the manufacturers.

Furthermore, in technical works one finds that detailed information is only given when the processes have been extant for a number of years—in other words, when they have become classic. This is not the case with the electro-metallurgy of aluminum, the evolution of which is as yet incomplete.

The minute details of manufacture are not likely

to interest the majority of readers, who expect to find in a monograph a history of the question and general information as to the manipulation and application of the metal.

2. My critic acknowledges the abundance of material I have collected, but he finds that "particularly in the latter portion of the book, the experimental results are presented in such form as to render almost *nil* the advantage of such a collection of facts, because the text is not supported by references sufficiently complete."

This last observation might be of significance, were it not for the fact that during seven years (1887–1894) I was occupied almost exclusively in the manufacture, the manipulation and the application of aluminum.

In so far as the author is concerned, the acquired experience might be considered sufficient, and the book satisfactory and complete, without the citation of references for the facts which were the results of the author's own observation and experiment. I have, nevertheless, cited a large number of investigators, both engineering and commercial, who have made contributions to the aluminum industry, and no omissions have been brought to my attention; the investigator who desires to carry his researches further should have recourse to the works of these authors.

3. Among the applications of aluminum, it is said that I have neglected to particularize two or

three which have rapidly developed during the last few years, such as: *the use of aluminum in lithography to replace the lithographic stone; the Ropeer-Edelmann process for the disargentation of lead; the use of powdered aluminum for colors in printing and painting.*

No doubt there are many other applications I have omitted to mention. This is but natural: the applications of aluminum are well-nigh innumerable, and now that its selling-price is hardly more than half a dollar per kg, and volume for volume, taking into consideration its lightness, it is found to be cheaper than most of the common metals—excepting, of course, iron, zinc, and lead—it would be very difficult to make a complete list. Nevertheless, the examples given by Mr. Haber are of interest, and I am glad to have been the occasion of mentioning them.

4. My statistics concerning the production of aluminum are nowhere near the true figures. While I have given, for 1900, a total production of 5,000 tons, Mr. Haber affirms that it amounts to 8,000 tons; other writers say 6,000 or 7,000 tons, others still maintain that the production of 8,000 tons was not attained until in 1902.

This uncertainty results from the fact that it is generally very difficult to obtain exact information from those interested, and this information, like that relative to the production, gives no details of the secrets of manufacture. Furthermore, it is

only possible to get the figures second-hand, given in round numbers or incidentally.

An interesting fact, for example, is the amount of energy necessary for the production of a kilogram of aluminum. Admittedly, this varies between 40 and 50 horse-power-hours; or, in other words, with two horse-power-days of 24 hours, a kilogram of aluminum can be produced.

With the total power of all the manufacturing establishments combined (see page 140), amounting to 61,000 horse-power, the entire daily output would be 30,500 kgs or 30.5 tons, and the entire annual production (360 days) would amount to 11,000 tons.

For the selling-price, it is said that my figures are too high.

It is admittedly a difficult matter to establish a fixed price even in the case of a long-established industry: it varies with the locality according to the ingredients needful for the manufacture, the skilfulness of administration, the continuity of labor, the price of labor, the cost of the reduction of the ore, etc.

The aluminum establishments are no exception to the rule; and it is impossible to give the cost of manufacture of the metal, except in general terms, for the very reason above given, that the figures vary according to the place of manufacture.

5. I have been criticised for not having devoted more than a very few pages to the Héroult and Hall processes, which are the only ones at present ap-

plied, while I have described at length and with complaisance the process I myself devised—a process which now possesses an interest merely historic, though my old establishment at St. Michel continues to operate, with no precise information as to the method employed.

After having passed in review a certain number of experiments which gave me no significant results, even in the laboratory, I have said that only the processes of Héroult, Hall and Minet have received the sanction of practice (page 78); and then I proceed to describe them.

If I have described my own process at some length, this is only because my process gave me the opportunity to study from the scientific point of view electrolysis by igneous fusion, and to establish the value of a certain number of constants, little known or undetermined before my researches (1887–1890).

I have at the same time established the regular formula of electrolytic reaction, that is to say, the relation between the current-elements, electromotive force, bath-resistance, and the intensity, and I have indicated the different factors that affect this formula in terms of the density of the current at the positive electrode (intensity per dc^2 of surface of the anode). Operating on a similar bath at different temperatures, I have determined that the counter-electromotive force diminishes in proportion as the temperature increases.

I have established the influence which the kind of cathode has on the quantity of metal deposited for a given quantity of current traversing the bath.

Finally, I have given the results of experiments showing that the liquid electrolyte which has been the subject of my observations was a *reversible electrolytic system* (page 100).

The bath studied by us was a mixture of 60 parts NaCl and 40 parts of Al_2F_6,6NaF (cryolite), with a mixture of Al_2O_3 and Al_2F_6 for alimentary substances.

Then I have observed that when one operates on baths of different composition, at least those with a base of cryolite, such as the baths used by Messrs. Héroult and Hall, where this salt is mixed with proportions more or less large of chlorides and alkaline fluorides and aluminum, one obtains for the counter-electromotive force values identical with that of my experimental bath, where the electrolytic reaction is the same in every case; and furthermore the current-elements satisfy the same regular formulas, according to the current-density at the positive electrode.

In the paragraphs treating of the Héroult and Hall processes, it only remained to make clear the points of difference between their methods and mine. Not knowing the exact formula for the baths employed by these engineers, I could do no more than to describe their apparatus. This is what I have done.

I might have said that besides the electrolytic system (comprising furnace and baths) adopted in the preparation (properly speaking) of aluminum, there exist devices for the preparation of electrodes, for instance, from the alimentary products Al_2O_3 and Al_2F_6, of which I have made no mention. To do so would have taken me too far from my subject, and it was a matter too subtle to handle.

It is interesting, nevertheless, to call attention to the recent experiments made by Mr. Hall, with the purpose of directly utilizing bauxite, partly transformed, in the preparation of pure aluminum.

I have myself made researches similar to those of Mr. Hall, at the St. Michel establishment (1892–1894) and at the laboratory of Mr. Le Verrier in Paris (1895–1896); and the results I have obtained have been the subject of communications made at about those dates. But this new method is at present being fully investigated, and to speak of it in detail without abundant technical information would be inopportune.

The Theoretical Part.

From the point of view of pure theory, Mr. Haber raises two questions: the one of secondary importance is that which I have first treated; the other, on the contrary, is a matter of the greatest interest, which I shall strive to elucidate, so far as the actual state of the art will permit.

1. It has been recognized for a long time that silicon has a deleterious effect on aluminum, notably on the resistance of this metal to the attacks of atmospheric and chemical agents.

Among other things, I had noticed that the plates or ingots of aluminum containing sufficiently large amounts of silicon were covered, at the end of a relatively short time, with a layer of white powder, rasping to the touch, and giving the sensation, when pressed between the fingers, of small grains of sand; I had concluded that this powder was composed of silica, without more exact observation, and I imagined that the silicious aluminum must be submitted to an interior working of which one of the effects was the displacement of the silicon, in proportion to the oxidation upon the surface (page 144).

It was a hypothesis purely gratuitous, and Mr. Haber professes his astonishment that I should have adhered to it, which amounts, he declares, to my saying that in case there is oxidation this would operate rather to the detriment of the aluminum than to the detriment of the silicon.

In reality, the heat of formation of the silica SiO_2 being equal to 45 cal. is weaker than the heat of formation of $\frac{1}{6}Al_2O_3$, which is 65.5 cal.; it follows that, of the silicon and the aluminum present, it is this last element which is the first to be oxidized. This order of oxidation is certain, as long as the mixture of aluminum and silicon is intimate; but in proportion as the aluminum is

oxidized, small grains of silicon are isolated from the entire mass of the metal, surrounded as they are by aluminum, and silicon in its turn is oxidized.

With this explanation it must also be admitted that the oxygen of the air, supposing that the metal is not absolutely homogeneous, attacking more actively certain parts of the surface, penetrates further and further into the interior of the mass, instead of the silicon making its way toward the surface.

Although this is true, from these various hypotheses it is certain that the silicious aluminum in the condition of plates somewhat thin, or of filaments, rapidly loses its mechanical properties. We have seen cases where, at the end of some months of exposure to the air, especially in the neighborhood of the sea, pieces formed of this aluminum had become very fragile; some of them crumbled into powder.

The same phenomenon of disintegration operates slowly in the case of light alloys of copper, even when exempt from silicon.

We believe that the light alloys of iron and of manganese are those which are disintegrated least rapidly, in contact with the air, and even with chemical agents.

2. I shall now mention the principal part of the criticisms of Mr. Haber—that which deals with electrolytic reactions and their continuity, by a rational alimentation of the bath in proportion to its decomposition.

The bath especially experimented with by us, and of which the composition is given above, corresponds to the formula $12NaCl + Al_2F_6, 6NaF$.

In proportion to its decomposition by the current, it is fed by a mixture of alumina and fluoride of aluminum: $nAl_2O_3 + Al_2F_6$, *with this condition, that* Al_2O_3 *should be raised in proportion as the temperature was lowered;* this last observation had not yet been made by me in the accounts of my previous studies, or at least it had not been presented in that form.

Experience has shown us that in the passage of the current events take place *as though, of all the compositions present, only* Al_2O_3 *disappeared,* whatever may be the hypothesis formulated as to the electrolytic reactions properly so-called and the local reactions; that is to say, the reactions do not interfere with the counter-electromotive force.

First Hypothesis.—In case one admits that Al_2O_3 dissolves in the bath and sinks, and that it is, of all the electrolytes present, the one that is attacked by the current—this is the hypothesis put forward by Héroult and Hall—the principal electrolytic is expressed by the relation

$$Al_2O_3 = Al_2 + O_3,$$

which becomes, when the quantity of current set in action equals 96,540 coulombs (chemical equivalent

of electricity), $\frac{1}{6}Al_2O_3 = \frac{1}{6}[Al_2 + O_3]$, the atomic weights of the elements which enter into reaction being taken with a single value and expressed in grammes. The aluminum goes to the cathode, the oxygen to the anode.

The oxygen in the nascent state attacks the carbon of the anode and forms carbon dioxide $O_2 + C = CO_2$. In fact the anodes are consumed proportionally to the amount of metal produced.

Second Hypothesis.—The electrolyte attacked by the current is the aluminum fluoride Al_2F_6; such is my hypothesis. Upon the passage of the current is produced the reaction

$$Al_2F_6 = Al_2 + F_6.$$

The aluminum goes to the cathode, the fluorine to the anode. Here are two hypotheses as to the rôle played by the fluorine.

(*a*) The fluorine displaces the oxygen of Al_2O_3 by the reaction $F_6 + Al_2O_3 = Al_2F_6 + O_3$; the aluminum fluoride is regenerated, and the nascent oxygen burns the carbon of the anode, forming CO_2: $O_2 + C = CO_2$.

(*b*) The fluorine, in the nascent state, combines with the carbon of the anode to form a tetrafluoride, $F_4 + C = CF_4$, and it produces between CF_4 and Al_2O_3, below or suspended in the bath, a double reaction, $3CF_4 + 2Al_2O_3 = 2Al_2F_6 + 3CO_2$, which regenerates the aluminum fluoride and burns the carbon of the anode.

The analysis of the phenomena which may take place in the various cases leads to the same conclusions as observation, that is to say, that whatever the hypothesis adopted, the only electrolyte which disappears from the bath is Al_2O_3, the aluminum precipitating itself at the cathode, the oxygen disengaging itself in the form of carbon dioxide. If, then, we proceed to a rational alimentation, that is to say if we feed the bath with quantities of Al_2O_3 proportional to the quantities of aluminum precipitated, the proportions of Al_2F_6 in the bath remain identical with themselves.

And if we add at the same time with the alumina certain proportions of Al_2F_6, it is to compensate for the very small quantities remaining of this fluoride, caused by the fact that in the hypothesis in which it constitutes the principal electrolyte it loses small quantities of CF_4.

Even in the hypothesis where the principal electrolyte is alumina, there is no doubt that fluoride of aluminum is decomposed at the same time. The heat of formation of Al_2F_6 being nearly that of Al_2O_3, with the result that, in every case, the alumina of alimentation should contain some proportions of aluminum fluoride. Here are the objections raised by Mr. Haber to this theory.

"The electrolyte recommended by Mr. Minet ($_{12}NaCl + Al_2F_6, 6NaF$) contains two anions, Cl and F, and two cations, Al and Na, all the anions as well as the cations in considerable proportions.

"According to the law of dynamics, the current discharges at the electrodes the materials of which the discharge requires the least effort; hence, in the present case: Cl at the anode; Al at the cathode. In this way the bath loses constantly Al_2Cl_6, and it is necessary to add to it this salt to keep constant the composition of the bath.

"M. Minet is a stranger to this point of view. He takes into consideration only the salts present, Al_2F_3, NaCl, NaF, without admitting of a change of ions, and then he thinks that it is Al_2F_6 which is first decomposed, as having the least heat of formation, etc.

"In this case the electromotive force should be 3.05, if one allows 70 calories for the heat of formation of $\frac{1}{6}Al_2F_6$, while experiment gives, according to the temperature, values of ε varying between 2.17 and 2.5 volts, which are very near that corresponding to $\frac{1}{6}Al_2Cl_6$."

At the time of my researches, and when the German edition had appeared, the heat of formation of $\frac{1}{6}Al_2F_6$ had not yet been determined by experiment. I had deduced the figure 70, by comparison between the heat of formation of the hologenic salts of aluminum and of potassium. But since then, M. Baud has established experimentally that this heat of formation is 83.17.

Furthermore, in his argument M. Haber has not taken account of the presence of alumina; but as the heat of formation of $\frac{1}{6}Al_2O_3$ is greater than that

of $\frac{1}{6}Al_2Cl_6$, this would not alter his conclusions in the least were not his argument radically incorrect, as we shall demonstrate.

Let us give first the heats of formation of the compounds present in the bath, and of those which might form whether by the exchange of ions or in the secondary reactions: these last may be either electrolytic or local.

Let us call C the heat of formation in the solid state of the chemical molecule expressed in grammes; T_c the heat of formation of the electrolytic molecule, that is to say of the chemical molecule taken with a single value of each one of the ions which compose it; e the electromotive force corresponding to T_c. C and T_c are expressed in great calories. e is given in function of T_c according to Thomson's rule: $e = T_c \times 0.0434$ volts.

	C		T_c	e
Al_2Cl_6	323.70	$\frac{1}{6}Al_2Cl_6$	53.95	2.34
Al_2O_3	393.	$\frac{1}{6}Al_2O_3$	65.50	2.84
Al_2F_6	499.	$\frac{1}{6}Al_2F_6$	83.17	3.61
NaCl	97.90	NaCl	97.90	4.25
NaF	110.80	NaF	110.80	4.81
CF_4	133.60	$\frac{1}{4}CF$	33.40	1.45
CO_2	97.6	$\frac{1}{4}CO^2$	24.40	1.06

M. Haber supposes, then, that in my bath *it is the chlorine which should appear at the anode*, while aluminum is deposited at the cathode, and as proof

in support he remarks that the counter-electromotive force which results from this reaction ($e = 2.34$) is precisely equal to those which I have found experimentally: 2.5 volts at 870°; 2.1 at 1,100°.

Since he cites in support of his contention my experimental results, I shall answer that in the first place *experiment* shows that not the least trace of chlorine is disengaged, when one electrolyzes a dissolved mixture of cryolite and alkaline chlorides. This is a fact conclusively proved, for we do not discover about the bath the slightest characteristic odor of chlorine; and if this halogen gave it off, the atmosphere round about would be absolutely unbreatheable.

I repeat, I have prepared from 1887 to 1894 about 100 tons of aluminum, with the bath I have indicated, and I have never noted in the atmosphere the least trace of chlorine.

We must then admit, in the present case, that the laws of dynamics may be satisfied, either that the sodium chloride has not suffered any dissociation, or that the relative values of the heats of formation of the electrolytes present are inverted; that is to say, that the heat of formation of Al_2Cl_6, which at the ordinary temperature is less than the heats of formation of Al_2F_6 and of Al_2O_3, is greater at the temperature of the operation; or rather, that the phenomenon takes place according to the indications which we give in the last paragraph.

According to the new order, it will not do to apply, except with great discretion, the laws of dynamics

to the dissolved electrolytes, if one takes as a base the heats of formation taken at 15°, and then works at temperatures of nearly 1,000°.

It is preferable to follow another method of analysis; for example, instead of seeking to establish, *a priori*, the counter-electromotive forces put in play, as functions of uncertain heats of formation, it is a surer method to calculate these counter-electromotive forces as functions of the regular formula, and to deduce from the values of e, thus determined, the reactions corresponding. It is thus that we shall proceed. We know that for the current-densities (intensity in amperes per sq. dm), varying between 2 and 100 amperes, the elements of the current satisfy the given regular formula $E = e + \rho I$ (page 94), in which E is the difference of potential taken at the electrodes; ρ the resistance of the bath; I the intensity of the current.

This formula is applicable not alone to the bath that we have more especially studied, but to every bath containing cryolite, even when one changes the proportions of the composition; it is verified, whether in the case of the Héroult bath constituted almost solely of cryolite and alumina, or in the case of Hall's baths, in which the cryolite is mixed with variable proportions of alkaline chlorides and fluorides and alumina.

In a word, in these different baths, it is *aluminum fluoride*, one of the constituent parts of the cryolite, or alumina which determines the counter-electromotive force, with all other secondary reactions.

Let us now glance at all the reactions which may take place: principal reaction, secondary electrolytic reactions, or purely local, according as we admit for the electrolytic principle Al_2F_6 or Al_2O_3.

Value of e deduced from the regular formula. — In the bath which has been the basis of our study the counter-electromotive force deduced from $[E = e + \rho I]$ varies with the temperature from 2.5 volts (temp .870°) to 2.17 volts (temp. 1,100°). We shall investigate now the reactions which correspond best to these values of e.

The principal electrolyte is Al_2O_3. — (*a*) *The secondary reaction is local.* The electrolytic reaction is reduced to the decomposition of $Al_2O_3 = Al_2 + O_3$; and for a single valence $\frac{1}{6}Al_2O_3 = \frac{1}{6}[Al_2 + O_3]$ $T_c = 65.5$ great calories.

$e_1 = T_c \times 0.0434 = 65.5 \times 0.0434 = 2.84$ volts, a value higher than that which gives the direct measure even for the lowest temperature 870°, where $e = 2.50$ volts.

It must, then, be admitted, if the case under discussion is indeed a real one, that we have taken for T_c a value too great; in other words that the heat of formation of Al_2O_3 diminishes in proportion as the temperature is increased. In fact, experience goes to show that the electromotive force, of which the value is proportional to the heat of formation of the electrolyte, rises from 2.50 volts at 870°, to 2.17 volts at 1,100°.

(*b*) *The secondary reaction is electrolytic.* It

should, then, re-enter into the calculation of the counter-electromotive force.

In the particular case, this secondary reaction is nothing but the oxidation of the carbon of the anode by the oxygen which originated at that anode in case (*a*).

$\frac{1}{4}(O_2+C)=\frac{1}{4}CO_2$, $T_c=24.4$, $e_2=1.06$. This reaction being exothermic, e_2 is a subtractive term. For the values of the counter-electromotive force desired, it amounts to

$$e=e_1-e_2=2.84-1.06=1.78 \text{ volt},$$

a value much less than that which gives the direct measure.

The principal electrolyte is Al_2F_6.—We follow the same method of analysis as in the preceding hypothesis.

(*a*) *The secondary reactions are local.*—The electrolytic reaction is reduced to $\frac{1}{6}Al_2F_6=\frac{1}{6}[Al_2+F_6]$, $T_c=83.17$, $e_1=3.61$ volts.

A value much higher than that of the direct measure, but at the same time with this principal reaction we know that there result from it secondary reactions which have the effect of fixing the fluorine while regenerating $\frac{1}{6}Al_2Fl_6$, and of burning the carbon of the anode. They may be produced in two different ways.

(*b*) *The secondary reactions are electrolytic.*

(1) First type of secondary reactions. The fluorine which orignates at the anode there meets

with alumina, in suspension or dissolved in the bath, it attacks this alumina; Al_2F_6 is regenerated, and the oxygen resulting from the decomposition of Al_2O_3 burns the carbon of the anode. So there result two secondary reactions, each of which, if they are electrolytic, furnishes a subtractive term for the calculation of e.

(1) $\frac{1}{6}F_6 + \frac{1}{6}Al_2O_3 = \frac{1}{6}O_3 + \frac{1}{6}Al_2F_6$,
$T_c = 83.17 - 65.5 = 17.67$, $e_2 = 0.77$,
$e' = e_1 - e_2 = 3.61 - 0.77 = 2.84$.

Same value of e as for the case where Al_2O_3 is the principal electrolyte, without secondary electrolytic reaction.

(2) $\frac{1}{4}O_2 + C = \frac{1}{4}CO_2$, $T_c = 24.4$, $e_3 = 1.06$,
$e = e_1 - e_2 - e_3 = 3.61 - 0.77 - 1.06 = 1.78$.

Again the same value as when alumina is the principal electrolyte, if one admits that the oxidation of the carbon of the anode is an electrolytic reaction.

(2) Second type of secondary reactions.

(*a*) The fluorine in the nascent state forms with the carbon of the anode a tetrafluoride (exothermic reaction).

$2F_6 + 3C = 3CF_4$, $\frac{1}{4}CF_4 = 33.40$,
$T_c = 33.40$, $e_2 = 1.45$.

The term e_2 being subtractive, for the value of the counter-electromotive force it amounts to

$$e' = e_1 - e_2 = 3.61 - 1.45 = 2.16.$$

Value very nearly that of the direct measure.

(*b*) The tetrafluorine attacks the aluminum; Al_2F_6 is regenerated.

$$\underset{(3\times 133.6)}{3CF_4} + \underset{(2\times 393)}{2Al_2O_3} = \left[\underset{3\times 97.6}{3CO_2} + \underset{(2\times 499)}{2Al_2F_6}\right] = 104 \text{ calories.}$$

This reaction is produced with a releasing of 104 calories, which, taken at a single valence, is $T_c = \frac{104}{2\times 6} = 8.67$ calories, $e_3 = 0.376$ volt. The term e_3 is subtractive.

$$e = e_1 - e_2 - e_3 = 1.78.$$

We return to the case where Al_2O_3 is the principal electrolyte, with a secondary electrolytic reaction.

To summarize: Of all the hypotheses which we have just passed in review that which appears to agree best with the direct measurement is the example (2) (*a*) for which the principal electrolyte is Al_2F_6, with one secondary reaction: an electrolytic reaction constituted by the formation of a tetrafluoride by the fluorine in the nascent state, and the carbon of the anode; the tetrafluoride of

carbon is released or produces a local reaction consisting of the double decomposition between the tetrafluoride liberated and Al_2O_3.

Whether there be truth or not in these hypotheses, one fact is undeniable, that when one electrolyzes a mixture of cryolite and sodium chloride with alumina in suspension or dissolved in the bath, the electrolytic reactions are explained by a disappearance of alumina, Al_2O_3.

Let us examine, finally, the hypothesis of the ion of chloride and of its discharge with the method employed above in the study of the electrolyte of Al_2F_6 and Al_2O_3.

The principal electrolyte is Al_2Cl_3.—At the temperature of the electrolysis, Al_2Cl_6 could not exist in the bath, in so far as definitely compounded, but we may express the discharge of the ions Cl_6 and Al_2 proceeded from a decomposition of Al_2Cl_6.

$$\tfrac{1}{6}Al_2Cl_6 = \tfrac{1}{6}(Al_2 + Cl_6),$$
$$T_c = 53.95,\ e_1 = 2.34.$$

This reaction could not be isolated, since we do not find any disengaging of chlorine. So we must admit that this halogen is newly determined by a secondary reaction, for example the effect upon Al_2O_3 of Cl in the presence of the carbon of the anode with the regeneration of Al_2Cl_6.

$$2Cl_6 + 2Al_2O_3 + 3C = 2Al_2Cl_6 + 3CO_2.$$

This double reaction produces a disengaging of heat, for the values of $T_c = 12.85$, $e_2 = 0.56$, from which $e = e_1 - e_2 = 2.36 - 0.56 = 1.78$ volt.

The electrolytic reactions are explained once more by a disappearance of Al_2O_3, a precipitation of Al at the cathode, and a releasing of CO_2 at the anode.

In all the cases studied, the composition of the bath is kept constant by a rational alimentation of aluminum.

Note that the hypothesis of the discharge of the ions Cl and Al at the electrodes—if one supposes that the secondary reaction is purely local—gives an electromotive force 2.34, very nearly the value found experimentally. This would be, then, a confirmation of a part of the argument of M. Haber, for if the phenomenon takes place thus, experience shows clearly that the bath does not become poor in Al_2Cl_6, as this physicist maintains, but in Al_2O_3 solely.

ALUMINUM IN THE UNITED STATES.

SUPPLEMENTARY NOTE BY THE TRANSLATOR.

It was a fortunate thing for the laurels of American metallurgical engineering development that the stalwart citizen of the Western Reserve, Edwin Cowles, then sixty-three years of age, leader of Republican movements, builder-up of the *Cleveland Leader*, and his two likely sons, Alfred and Eugene H. Cowles, should in 1883 have purchased a New Mexican mine on the Pecos River whose output consisted of extremely refractory zinc ores. Both of the sons were ingenious and resourceful. Eugene added to a practical knowledge of the difficulties of making solid-steel castings the training of a writer on his father's paper and the executive experience of the management of an electric lighting plant at a time when difficulties to be overcome were those of ignorance on the part of a public and uncertainty as to the engineering results in the output of power and lighting plants. Alfred had followed more closely the lines of study and experimental research. He attained distinction in his university life at Cornell. His mind was constantly active and with a strong tendency to at once submit to experiment the ideas which its fertility suggested.

In this father and sons was present that combination of social and business prominence, mental force, and courage of conviction necessary to overcome that inertia which had up to this time relegated the possibilities of aluminum to a use supposedly remote in its development.

The testimony of these gentlemen in the long-continued patent litigation forms the most important source of history in this connection. A sketch in the note-book of Mr. Eugene H. Cowles dated June, 1883, and bearing the title "Proposed Electric Furnace for working Pecos Ores," contains the essentials of the later patented forms for the smelting of aluminum alloys. The mass of mixed ores for reduction by incandescent heat, the posing inclined carbon terminals, the vents at the tops of the furnace, and the tap-hole for withdrawing the molten charge, are all present.

The Pecos River had a fall of 75 feet to the mile. The absence of fuel made the application of electrical heat a thing to be considered. At that time the alternating current was in a struggling infancy. The art of building dynamos which should possess enormous amperage output had not been developed. When, therefore, in 1886 the Brush Company of Cleveland, acting under the enthusiastic inspiration of the Cowles Brothers' proximity, built the largest direct-current generator which the world had seen, and which produced thirty-four hundred amperes at sixty-eight volts, it was called, for distinction, "The Colossus." The output of this machine was consequently about 230 kw.—a small beginning

as compared with 13,000 kw., the estimated energy used in the American production of aluminum for current American manufacture.

The transition from the application of the electrical furnace for the smelting of refractory zinc ores to the production of the silver made from clay and the copies of gold known as aluminum bronze was immediate. Aluminum in its pure state had been the dream of chemists. It was so rare that service trials were practically unknown. If it could be obtained, navigation would be immensely profited, for ships would be light and tonnage vastly increased. Railway trains would be imperishable and track wearage reduced to a minimum. Aerial navigation would be expedited. Military accoutrements, fixed ammunition, and guns would be lightened. Innumerable uses would spring up for this wonderful metal which was popularly described as being as strong as steel and but little heavier than wood. The presence of its ore in the earth's crust in large excess even of that of iron itself would furnish inexhaustible sources for the new Aluminum Age. Practical metallurgists were silent as to the actual use of aluminum, for it was not in their experience. It is no wonder, therefore, that the scheme of "harnessing Niagara" should have appealed vividly to the Cowles family and their associated friends. The Pecos River had enabled them to consider the use of water-power as a necessity. The slightest calculation showed that the units of heat necessary for metallurgical operations with aluminum ores required enormous power.

The proximity of Cleveland, with the Brush works, where machines could be built, to the then existing tail-race of the Niagara overflow at Lockport, New York, suggested a combination of agencies already established which would most quickly lead to electrical smelting on the large scale necessary. We now know that aluminum has most serious drawbacks to the uses predicted for it by its early sanguine exploiters. No one at that time could have foreseen that its largest use would be as a reagent in the manufacture of steel, and that only in combination with other metals or even with such apparently unrelated substances as phosphorus does it get structural solidity and machining value. It is yet to be seen what the surprising development of aluminum in aluminothermics will be, but one thing is evident, that the freely disseminated literature issued by the Cowles Company in its early history, while it helped the greatest single advance which metallurgy has made, was based upon a largely erroneous view as to the true application of its results. The Cowleses were not the people who hid lights under bushels. The success of the Cowles furnaces at Lockport was at once followed by the description of their results in the prominent engineering journals of Europe, and with characteristic energy the establishment of plants with adequate financial backing in England. Most important was the world-wide attention drawn to the possibilities of electric smelting, and the establishment of the very many works M. Minet has outlined in this volume.

By far the most important of these outgrowing developments in the aluminum industry, so far as America is concerned, was that arising from the work of Charles M. Hall of Oberlin, Ohio, who was an attaché of the Cowles Lockport works in the early years of its history. The Cowles process had failed to produce pure aluminum unalloyed with other metals, as a practical outcome from the Cowles furnaces. Hall, after leaving the Cowles works, embodied a practical method for the production of pure aluminum by electrolysis in five patents issued in 1889. These patents describe the electrolysis of alumina while held in solution in a molten bath of cryolite to which various other ingredients have been added. Very much has been written as to the actual reaction taking place in the Hall process. Dr. Ahrens * sums up the reaction when he describes the Deville work in 1859 in the following words which I have translated: "Deville thereupon arrived at the conception to investigate it [i.e., the preparation of Al by electrolysis] with other compounds; in a short paper of the year 1859 he described a process which rested upon the principle of decomposing molten cryolite by the electric current, and regenerating the cryolite by the use of anodes containing Al_2O_3.

"Doubtless Deville was, in this, reverting to an analogy with the chemical preparation of Al; just as by the effect of Cl upon a mixture of Al_2O_3 and

* Handbuch der Elektrochemie, von Dr. Felix B. Ahrens, Stuttgart, 1903 [2d Edition], p. 505.

carbon, $AlCl_3$ and CO are generated, so should the Fl set free by the electrolysis of cryolite act upon the mixture of Al_2O_3 and carbon of the anode, and in this way regenerate the AlF_3, which is used by the electrolysis.

"The process of Deville failed, first, because Deville conducted from outside the heat necessary for the melting of the cryolite, which the decomposition-vessels could not stand; then, however, it failed at the easily crumbling carbon—Al_2O_3 anodes."

It will thus be seen that the essential point of the solution of alumina in molten cryolite was practically tried by Deville, and that he himself offered the explanation that it was the cryolite itself which was electrolyzed, while the alumina regenerated the cryolite instead of being itself electrolyzed. This distinction has an important bearing upon the patent situation when the question of external heating is involved. The electrolysis of alumina in solution would require not more than one-half the voltage given as the voltage necessary in the practical operation of the Hall patents mentioned. If the required voltage for the electrolysis of alumina only were allowed, in the writer's opinion it would be impossible to have operated the Hall processes, even though abundant external heat was supplied from non-electrical sources.

The Hall process under the excellent management of the Pittsburg Reduction Company has steadily developed until now good judges base the annual output in America alone at about

15,000,000 lbs. of pure aluminum annually.* This company, by wisely refraining from charging unreasonable prices by virtue of its monopoly, has made it possible to use aluminum as a staple at a fixed price: a condition which is primary to extended uses in metallurgical engineering. Although protected by a duty and thus presenting one of those unexpected conditions under our tariff by which competition is completely suppressed, the cost to the user has always been just. For some years there existed a continuous litigation between the Pittsburg Reduction Company and the Cowles Company, which latter was enjoined from utilizing what seemed to them to be a natural development of their process. Recently † the Cowles Company have received at least something of poetic justice, for it has been decided by the courts that the Hall processes, having always been operated under the influence of heat generated by the current which is used also for the electrolysis, are subject to a patent controlled by the Cowles Company issued to Charles S. Bradley, in which the combination of the electrolyzing and internal heating current is protected. Mr. Alfred H. Cowles survives his father and brother, and alone witnesses this somewhat tardy justice. Meanwhile our knowledge of the metallurgy of aluminum has received accessions which make more possible the realization of the predictions of twenty years ago.

*See a description of the Niagara Falls plant, by J. W. Richards, *Electrochemist and Metallurgist*, Oct., 1902, p. 49.

†October, 1903.

For foundry purposes it is taking its place with other white metals and is seldom used alone. Its alloys with metals of its own class, such as magnesium and zinc, are rapidly attaining commercial importance where lightness is the desideratum. With steel a newly discovered but highly significant use has been found affecting the magnetic properties concerned in dynamo and transformer construction.

With reference to copper and brass and the finest of all copper alloys, the aluminum bronzes or aluminides of copper, obstructions of an unexpected nature have been met. The specific heat of aluminum, its latent heat of fusion,* and the avidity with which it seizes upon iron and silicon as impurities when in a state of fusion, have been occult reasons operating against its manufacture in works regularly devoted to brass and copper. So strongly entrenched are the ideas of the brass and copper melters regarding the behavior of any white metal like aluminum—ideas borrowed from the behavior of tin and zinc at melting temperatures—that it is all but impossible to secure technically proper treatment for the copper alloys of aluminum. This has led to the inaccurate naming of trade materials, so that it is now not possible to know from a simple announcement in trade circulars whether a metal designated as aluminum bronze may not contain zinc, tin, or even lead as well.

* Total number of calories to melt Cu=162, Al=258.3, Fe=370, Au=50.9, Ag=89.2, Pb=15.6, Zinc=67.8. (From *Electrochemical and Metallurgical Industry*, July, 1905, J. W. Richards.)

There is a tendency on the part of metallurgical engineers to depart from the production of aluminum from its oxide. Bauxite is limited in amount, and for any large production the question of freight charges or propinquity of water-power or the two together is a controlling factor. The presence of large bodies of excellent clays and the very high efficiency recently attained in the modern types of large-unit gas-engines promise in the not too distant future to completely change the sources and methods of production of metallic aluminum.

Much has been written upon the uses of aluminum for electrical conductors. It is generally assumed that for equal cross-sections we can now reach a two-thirds conductivity of copper. It is not improbable that this may be largely increased, since so far we have not been able to manufacture aluminum of a corresponding purity with copper. If the same kind and amount of impurity were introduced into our present good copper as now exists in our best aluminum, the conductivity of the copper would surely be lowered more than ten and possibly more than fifteen per cent. In Appendix A the conductivity tables prepared by the Pittsburg Reduction Company are given for convenient reference. In Appendix B is given a list of the more important aluminum patents issued by the U. S. Patent Office in recent years. In Appendix C is a tabular view of the output of aluminum in the United States, and in Appendix D the cost per pound of aluminum.

ALUMINUM WORKS IN AMERICA, 1903.*

Name.	Location.	Horse-power.		Process.	Capital.
		Available.	In Use.		
Pittsburg Red. Co.	Niagara Falls.....	.....	14,000	Hall	$1,600,000
" " "	" "	.....		"	
" " "	Massena Springs, N. Y.	1,200	.....	"	
(Royal Al. Co.)...	Shawenegan Falls, Quebec, Canada...	6,000	5,000	"	

* From The Production of Aluminum and Bauxite, 1903. By Joseph Struthers, Washington, Government Printing Office, 1904. (Extract from Mineral Resources of the United States.)

APPENDIX A.

TABLE OF RESISTANCES OF PURE ALUMINUM WIRE.*

Conductivity 62 in. the Matthiessen Standard Scale. Pure aluminum weighs 167.111 pounds per cubic foot.

Am. Gauge, B. & S. No.	Resistances at 70° F.				Log d^2.	Log R.
	R Ohms per 1000 Feet.	Ohms per Mile.	Feet per Ohm.	Ohms per Pound.		
0000	.07904	.41730	12652.	.00040985	5.325516	$\bar{2}$.897847
000	.09966	.52623	10034.	.00065102	5.224808	$\bar{2}$.998521
00	.12569	.66362	7956.	.0010364	5.124102	$\bar{1}$.099301
0	.15849	.83684	6310.	.0016479	5.023394	$\bar{1}$.200002
1	.19982	1.0552	5005.	.0026194	4.922688	$\bar{1}$.300639
2	.25200	1.3305	3968.	.0041656	4.821980	$\bar{1}$.401401
3	.31778	1.6779	3147.	.0066250	4.721274	$\bar{1}$.502127
4	.40067	2.1156	2496.	.010531	4.620566	$\bar{1}$.602787
5	.50526	2.6679	1975.	.016749	4.519860	$\bar{1}$.703515
6	.63720	3.3687	1569.	.026628	4.419152	$\bar{1}$.804276
7	.80350	4.2425	1245.	.042335	4.318446	$\bar{1}$.904986
8	1.0131	5.3498	987.0	.067318	4.217738	0.005652
9	1.2773	6.7442	783.0	.10710	4.117030	0.106293
10	1.6111	8.5065	620.8	.17028	4.016324	0.207122
11	2.0312	10.723	492.4	.27061	3.915616	0.307753
12	2.5615	13.525	390.5	.43040	3.814910	0.408494
13	3.2300	17.055	309.6	.68437	3.714202	0.509203
14	4.0724	21.502	245.6	1.0877	3.613496	0.609850
15	5.1354	27.114	194.8	1.7308	3.513788	0.710574
16	6.4755	34.190	154.4	2.7505	3.412082	0.811273
17	8.1670	43.124	122.5	4.3746	3.311374	0.912063
18	10.300	54.388	97.10	6.9590	3.210668	1.012837
19	12.985	68.564	77.05	11.070	3.109960	1.113442
20	16.381	86.500	61.06	17.595	3.009254	1.214340
21	20.649	109.02	48.43	27.971	2.908546	1.314899
22	26.025	137.42	38.44	44.450	2.807838	1.415391
23	32.830	173.35	30.45	70.700	2.707132	1.516271
24	41.400	218.60	24.16	112.43	2.606424	1.617000
25	52.200	275.61	19.16	178.78	2.505718	1.717671
26	65.856	347.70	15.19	284.36	2.405010	1.818595
27	83.010	438.32	12.05	452.62	2.304304	1.919130
28	104.67	552.64	9.55	718.95	2.203596	2.019822
29	132.00	697.01	7.58	1142.9	2.102890	2.120574
30	166.43	878.80	6.01	1817.2	2.002182	2.221232
31	209.85	1108.0	4.77	2888.0	1.901476	2.321909
32	264.68	1397.6	3.78	4595.5	1.800768	2.422721
33	333.68	1760.2	3.00	7302.0	1.700060	2.523330
34	420.87	2222.2	2.38	11627.	1.599354	2.624148
35	530.60	2801.8	1.88	18440.	1.498646	2.724767
36	669.00	3532.5	1.50	29352.	1.397940	2.825426
37	843.46	4453.0	1.19	46600.	1.297234	2.926064
38	1064.0	5618.0	.95	74240.	1.196526	3.026942
39	1341.2	7082.0	.75	118070.	1.095820	3.127494
40	1691.1	8930.0	.59	187700.	05.99112	3.228169

* Calculated on the basis of Dr. Matthiessen's standard, viz.: The resistance of a pure soft copper wire 1 meter long, having a weight of 1 gram = .141729 International Ohm at 0° C.

(From Aluminum for Electrical Conductors, The Pittsburgh Reduction Co. 1903.)

APPENDIX B.

LIST OF U. S. ALUMINUM PATENTS.

Date.	No.	Name.	Patent.
1881, July 12	244234	Paget-Higgs	Electrolyzes cryolite and borax.
1885, April 7	315266	Moses G. Farmer	Electrolyzes aluminum, chlorides, fluorides, etc.
1887, May 3	362441	Richard Gratzel	Reduces by magnesium *et al.* and electrolysis.
1888, Aug. 14	387876	Paul Heroult	Electrolyzes alumina.
1889, April 2	400664	Charles M. Hall	" "
" "	400665	" " "	" "
" "	400666	" " "	" "
" "	400667	" " "	" "
" "	400766	" " "	" "
1891, Dec. 8	464933	Charles S. Bradley	Melts with electrolyzing current.
1892, Jan. 5	466460	T. A. Edison	Electrolyzes al. chloride.
1892, Feb. 2	468148	Charles S. Bradley	Melts with electrolyzing current.
1892, April 19	473118	Paul Heroult	Electrolyzes alumina.
1892, April 26	473866	Charles S. Bradley	Electrolysis with blast flame.
1892, June 7	476256	M. Emme	Electrolyzes alumina.
1892, June 14	476914	Bernard Bros.	Electrolyzes cryolite and sodium fluoride.
1893, Aug. 22	503929	Joseph B. Hall	Electrolyzes alumina.
1894, Jan. 16	512801	Willard E. Case	Electrolyzes al. sulphate and calcium fluoride.
" "	512802	" " "	Electrolyzes al. sulphate and calcium fluoride.
1894, Oct. 23	527846	Waldo and Gooch	Electrolysis al. compounds.
" "	527847	Gooch and Waldo	" " "
" "	527848	" " "	" " "
" "	527849	" " "	" " "
" "	527850	" " "	" " "
" "	527851	" " "	" " "
1894, Oct. 30	528365	" " "	" " "
1896, June 23	562785	H. F. D. Schwan	Electrolysis aluminous minerals
1897, March 9	578633	F. A. Gooch	Electrolysis al. compounds.
1899, Aug. 15	631253	" " "	" " "
1901, April 30	673364	W. Hoopes	Purifies aluminum electrolytically.
1902, Dec.	715625	Taddei, G	Electrolysis. Decomposes NaCl at high temperature.
1903	732410	Homan	Manufacture of silicon and al. from silicates of alumina.
1904	763479	Gin, G.	Electrolyzes $Al_2S_3 . 3Na_2S$ at 850° C.
1904, May 24	760554	Onda, Myagoro.	Manufacture of sulphides of aluminum and alloys of al.
1904, Nov. 15	775060	Blackmore, H. S.	Electrolyzes al. oxide in combination.

APPENDIX C.

TABLE * SHOWING THE OUTPUT OF ALUMINUM IN THE UNITED STATES, 1883–1904, WITH CURRENT MARKET PRICES.

Year.	Quantity, Pounds.	Value, Dollars.
1883	83	
1884	150	
1885	283	
1886	3,000	
1887	18,000	59,000
1888	19,000	65,000
1889	47,468 (incl. alloys)	97,335
1890	61,281 (incl. alloys)	61,281
1891	150,000 (incl. alloys)	100,000
1892	259,885	172,824
1893	339,629	266,903
1894	550,000	316,250
1895	920,000	464,600
1896	1,300,000	520,000
1897	4,000,000	1,500,000
1898	5,200,000	1,716,000
1899	6,500,000	1,716,000
1900†	7,150,000	1,920,000
1901	7,150,000	2,238,000
1902	7,300,000	2,284,590
1903	7,500,000	2,284,900
1904	8,600,000‡	2,477,900

* Based on the Report of the Department of the Interior, United States Geological Survey, Division of Mining and Mineral Resources: Mineral Products of the United States, Washington.

† Statistics from 1900 to 1903 not forthcoming from manufacturers. Obviously the figures are much too low.—(Translator's note.)

‡ Too low. Probably 10,000,000.—Translator.

ALUMINUM PRODUCTION IN THE UNITED STATES.

Based on Neumann: *Die Metalle.* Halle: Wilhelm Knapp, 1904.

	Pounds Avoirdupois.		Pounds Avoirdupois.
1882	0	1888	19,000
1883	90	1889	48,000
1884	150	1890	60,000
1885	700	1891	170,000
1886	6,600	1892	290,000
1887	18,000		

APPENDIX D.

PRICE OF ALUMINUM.

	M. per kg. (wholesale, on Continent).	Cents per lb. (wholesale, on Continent).
1854	2400	259.20
1855	1000	108.00
1856	300	32.90
1857	240	25.92
1859	160	17.38
1864	160	17.38
1874	160	17.38
1878	105	11.34
1884	82	8.86
1885	74	7.99
1886	70	7.56
1888	44	4.75
1889	38	4.10
1890 Feb.	27.60	2.98
1890 Sept.	15.20	1.64
1891 Feb.	12	1.30
1891 July.	8	.86
1891 Nov.	5	.54
1892	5	.54
1893	5	.54
1894	4	.43
1895	3	.32
1896	2.60	.28
1897	2.50	.27
1898	2.20	.24
1899	2.20	.24
1900	2	.22
1901	2	.22

[1854–89. Prices by Deville process, various establishments. 1890–1901. Prices Electrolytic Al. from Neuhausen, Metallgesellschaft Frankfurt.—From *Die Metalle*, von Dr. Bukhard Neumann. Halle, Wilhelm Knapp, 1904.]

1902 (Pitts. Red. Co.), 31–37 cts. per lb.; 1903 (Pitts. Red. Co.), 1904, 1905, ditto.

U. S. Customs Duties as follows (July 1, 1902):

Aluminum alloys	8 cts. (lb.)
" articles	45 per cent
" crude	8 cts. (lb.)
" plates, sheets, bars, or rods	13 cts. (lb.)

LIST OF A FEW IMPORTANT TREATISES AND MEMOIRS ON ALUMINUM.

(By the Translator.)

1859. Deville, H. St. C. L'Aluminium; ses propriétés, sa fabrication et ses applications. Paris. 8°.

1873. Biedermann, R. Aluminium und Aluminium-Verbindungen. Vienna Univ. Exhibition, 1873; German Comm. Band III. Abt. I. Hälfte I. 1875. 8°.

1873. Wurtz, C. A. Ueber die Fabrikation des Aluminiums. Vienna Univ. Exhibition, 1873; German Comm. Amthicher-Bericht. Band III. Abt. I. Hälfte I. 1875. 8°.

1874. Tissier, C. and A. Guide de la recherche, de l'extraction et de la fabrication d'Aluminium, et des métaux alcalins. Nouv. éd. Paris. 12°.

1884. Margottet, J. Aluminium. *Fremy, E.* Encyclopédie chimique. Tome III. fasc. 4. 8°.

1885. Wierzinski, S. Die Fabrikation des Aluminiums und des Alkali-Metalle. Vienna. sm. 8°.

1887. Richards, J. W. Aluminium: its history, occurrence, properties, metallurgy, and applications, including its alloys. Philadelphia. 8°. [2d and 3d Editions since, in 1890 and 1896.]

1888. Naccari: Ueber die Specifischen Wärme einiger Metalle. Beiblatter zu den Annalen der Physik. XII, p. 326.

1890. Wickersheimer. L'aluminium et ses alliages. *Fremy, E.* Encyclopédie chimique.

1892. Le Verrier, U. Études sur l'aluminium. Conservatoire des Arts et Métiers. Annales. 2e Ser. Tome IV.

[1893–97.] Minet, A. L'aluminium. [I.] Fabrication, emploi. [II.] Alliages, emplois récents. Paris, 2 vols., sm. 8°.

1900. March 3. Aluminum Conductivity Tables, etc., in L'Eclairage Electrique, XXII.

1900. Methoden und Resultate der Untersuchung des Aluminiums und seiner Abkömmlinge. *Zurich: Schweizerische Polytechnicum, Austalt zur Prüfung von Baumaterialen.* Mittheilungen. Heft IX. 8°.—Zusammengestellt von Prof. L. Tetmajer.

1902. Feb. The Physical Properties of Certain Aluminium Alloys, and Some Notes on Aluminium Conductors. Ernest Wilson. Jour. Inst. E. E., London, E. and F. N. Spon.

1903. Moissonier, P. L'aluminium, ses proprietés, ses applications. Paris. 8°. (Gauthier-Villars.)

1903. Winteler, F. Die Aluminium-Industrie. Braunschweig F. Vieweg und Sohn.

1904. Borchers, W. Electric Smelting and Refining,—*Aluminum*,—Part II, p. 93. (Trans. by W. G. McMillan.) Lond. and Phila.

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